High-temperature moment-volume instability and anti-Invar of γ-Fe

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(Received 11 June 1993)

Just as Invar describes a system with a smaller than "normal" thermal expansion in a magnetically ordered state, a system with a larger than "normal" thermal expansion in a magnetically disordered state can be described as anti-Invar. Anti-Invar had been already observed in a number of fcc binary and ternary alloys of the 3*d* series well above magnetic ordering temperatures. While Invar is characterized by low-temperature moment-volume instabilities, anti-Invar signifies the importance of high-temperature moment-volume instabilities. Both are closely related to the ground-state properties. We have reinvestigated the $Fe_{50}Ni_xMn_{50-x}$ alloy system and confirmed the existence of enhanced thermal expansion at high temperatures, i.e., the anti-Invar effect. With the aid of the thermal properties of these alloys we deduce the temperature dependence of the thermal expansion of γ-Fe and show its anti-Invar nature. The enhanced volume expansion in γ-Fe amounts to 2.8%, which is larger than the Invar effect (2%) in $Fe_{65}Ni_{35}$.

I. INTRODUCTION

It is well established that the magnetic moment in 3*d* fcc metals and alloys is not necessarily a continuously varying quantity with atomic size, but can change discontinuously in the vicinity of a critical volume, thus exhibiting a moment-volume (MV) instability.[1-3] Total-energy calculations have predicted the existence of these instabilities in the Invar systems Fe-Ni and Fe-Pt, as well as the elements, e.g., fcc Mn, fcc Fe, and bcc Cr.[4-7]

In Invar alloys, the states for volumes larger and smaller than the critical volume are known as "high-spin high-volume" (HS) and "low-spin low-volume" (LS) states, respectively, and are separated by an energy difference of a few mRy. Evidence for the existence of these instabilities has also been given experimentally by low-temperature Mössbauer experiments under pressure on Fe-Ni and Fe-Pt Invar alloys.[8] The ground-state properties of Invar alloys are, therefore, to a large extent well understood. On the other hand, however, understanding the finite-temperature properties of these materials is more complex, for it requires the description of the temperature evolution of the MV instabilities. Finite-temperature theories developed by introducing fluctuations to the calculated ground-state properties and using Landau formalism or a Debye model have given substantial insight into the problem,[9-12] but a theory describing the thermal evolution of the electronic structure, which should lead to a complete understanding of the thermal properties of Invar, has not been established.

Despite the complexities, a simple but valid picture of the Invar effect can be acquired by boiling down the details of the temperature evolution of the instabilities to a thermal activation process between the HS and LS states, separated effectively by an energy difference $\Delta\varepsilon$. This simplied picture actually reminds one of the earlier Weiss model used in describing the Invar effect.[13] However, the existence of the HS and LS states no longer has to be hypothesized, and the model can now be grounded on the results of total-energy calculations and experiments, which yield the existence of these states. Yet, the physical nature of the excitations between these states is still a subject of intensive research.[7] It is assumed that the energetically higher small-volume LS state is progressively occupied with increasing temperature at the expense of the energetically lower-lying large-volume HS state, leading to a volume contraction which compensates the "normal" lattice thermal expansion. Although this model is still phenomenological in nature and does not give a first-principles account of finite-temperature properties, it has proved useful in describing the thermal behavior of Invar-type alloys.[13-16]

In contrast to, e.g., Fe-Ni Invar, where the ground state is the large-volume ferromagnetic (FM) HS state, calculations for γ-Fe have shown that the ground state is a low-volume antiferromagnetic (AF) state separated from the energetically higher-lying large-volume FM HS state.[5,17] The existence of the AF ground state and the FM HS state has been verified by Mössbauer experiments on γ-Fe precipitates in Cu and Cu-Al,[18] and on γ-Fe thin films grown on Cu_3Au.[19] The hyperfine field[18] shows a stepwise increase at a critical volume, $V_c = 12.05$ nm^3, on going from the small-volume AF state to the large-volume HS state. Furthermore, Mössbauer[20,21] and neutron-diffraction studies[22] on γ-Fe precipitates in Cu reveal AF order with $T_N \simeq 70$ K and a magnetic moment of $0.5\mu_B$, thereby confirming recent calculations.[17] Therefore, the ground-state properties of γ-Fe are well understood. However, an understanding of the properties of γ-Fe at finite temperature is still lacking.

0163-1829/94/49(9)/6012(6)/$06.00

To better understand the thermal properties of γ-Fe we first look at its behavior within the stability range of 1184 K $< T <$ 1665 K. Within this range, where γ-Fe is paramagnetic, the measured properties are already anomalous. Figure 1 shows the thermal-expansion coefficient of γ-Fe as a function of temperature reduced to the melting point $\alpha(T/T_m)$, in comparison to other fcc elements with no MV instabilities.[23,24] While $\alpha(T/T_m)$ of fcc elements like Cu, Ni, Pd, and Pt shows normal Grüneisen behavior, $\alpha(T/T_m)$ of γ-Fe in the stability range is almost temperature independent. The anomalous nature in this region is also seen in the fact that neutron-diffraction experiments on γ-Fe yield predominantly ferromagnetic (FM) coupling with a magnetic moment of about $1.0\mu_B$, whereas the temperature dependence of the inverse susceptibility leads to a negative paramagnetic Curie temperature calling for AF coupling.[25,26] Furthermore, it is known that the specific heat and the pressure dependence of the resistivity in the stability range of γ-Fe cannot be understood in terms of standard methods of analysis.[14] Above all, the AF coupling in the ground state must transform into FM coupling at some temperature or a temperature interval below the stability region.

Thus, to extract information on γ-Fe outside the stability range, we look at the evolution of the atomic volume V_a with temperature. V_a is a quantity which is directly related to MV instabilities. Its temperature dependence, therefore, holds information involving the thermal evolution of these instabilities. We assume that an activation process similar to that used to describe the thermal properties of Invar should also be capable of describing the thermal properties of γ-Fe. Since in γ-Fe, the HS state is the energetically higher-lying state, one can expect the opposite to what happens in Invar: with increasing temperature, a thermal population of the large-volume HS state at the expense of the low-volume AF state. This should lead to an enhancement in the volume expansion, hence "anti-Invar." In order to construct $V_a(T)$ for γ-Fe, we use the two-level system formalism, but stress that we do not extract from this the physical nature of the excitations. It is used because at present there is no theory describing satisfactorily MV instabilities at finite temperatures in magnetically disordered systems.

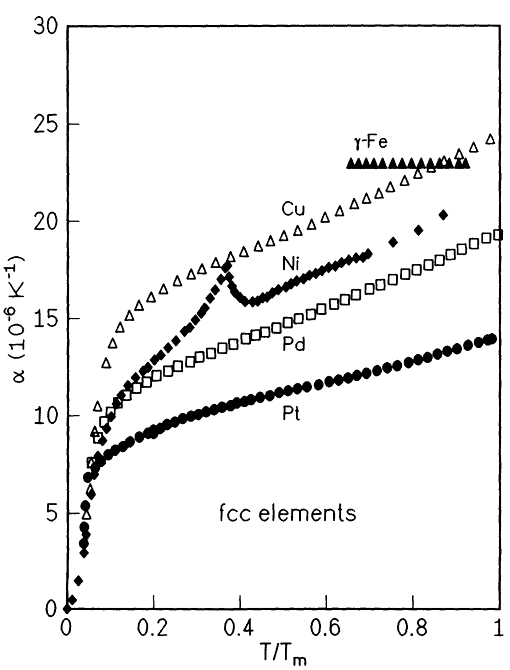

FIG. 1. $\alpha(T/T_m)$ of γ-Fe compared to those of selected fcc elements. $\alpha(T)$ of γ-Fe is nearly constant and deviates from the general trend in the elements.

Enhanced volume expansions at high temperatures in fcc Fe alloys had been observed in Fe-Ni, Fe-Ni-Mn, Fe-Mn-Co, etc.[15] A modified Weiss model had been successful in describing this behavior, as well as the high-temperature enhancements in other quantities such as the specific heat and the electrical resistivity.[15,27–31] However, at the time, high-temperature properties of these alloys were neither attributed to MV instabilities nor had they been used to understand the anti-Invar nature of γ-Fe.

The present study has, thus, two objectives. First, to reinvestigate fcc Fe alloys in a stability range where they show the anti-Invar effect and second, to examine, in view of the high-temperature properties of these alloys, the thermal behavior of the atomic volume of γ-Fe outside the stability range. We will show in the complete temperature range that γ-Fe has anti-Invar nature.

II. HIGH-TEMPERATURE $\alpha(T)$ OF $Fe_{50}Ni_xMn_{100-x}$

To confirm the existence of the high-temperature enhancements in various physical parameters in $Fe_{50}Ni_xMn_{100-x}$, we have remeasured the thermal expansion. Measurements were carried out on three cylindrical samples of 6 mm diameter and 7 mm length with $x=27$, 32, and 34 at. % from the same batch used in earlier experiments.[15] At these concentrations, the samples have AF, reentrant spin-glass (RSG) and FM ground states, respectively.

Two separate capacitive dilatometers are used to cover the temperature ranges 4.2–300 K and 300–1000 K. The low-temperature dilatometer is a conventional relative-copper-capacitance-cell with the sample thermally coupled to the cell. In the high-temperature dilatometer, the sample is situated in a variable temperature oven, and is mechanically coupled to a fixed temperature (308±0.001 K) capacitance cell via a quartz push rod. Measurements were taken in steps of ~2 K at thermal equilibrium at which the temperature is stabilized to ±1 mK for 4.2 K $< T <$ 300 K, and ±10–50 mK for 300 K $< T <$ 1000 K. The coefficient of thermal expansion is calculated as the slope of consecutive data.

$\alpha(T)$ of the alloys along with that of Ni are shown in Fig. 2. For all three samples, the high-temperature behavior far above the respective magnetic ordering temperatures is anomalous, specifically in comparison to Ni. These fcc alloys show a broad maximum in $\alpha(T)$, i.e., the anti-Invar effect, in excellent agreement with earlier data on similar samples.[27] In the following, we will deduce that γ-Fe has a similar behavior in $\alpha(T)$.

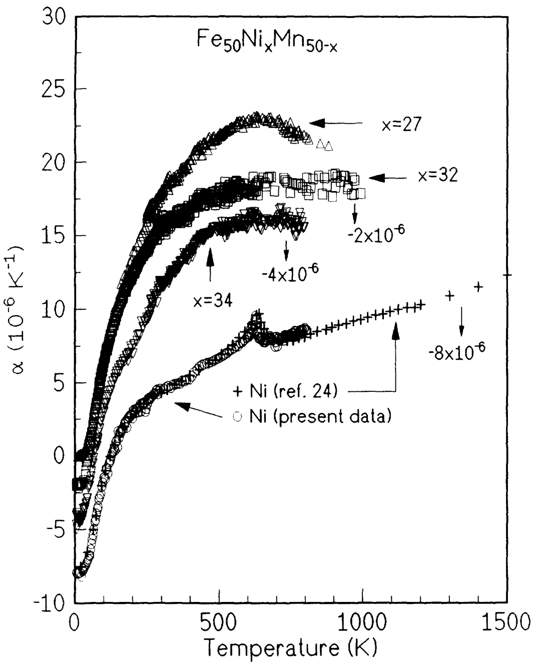

FIG. 2. $\alpha(T)$ of $Fe_{50}Ni_xMn_{50-x}$ compared to Ni. The data are shifted for clarity by amounts shown on the curves. The high-temperature data of the alloys deviate from Grüneisen behavior. $T_N(x=27)=140$ K, $T_C(x=34)=240$ K.

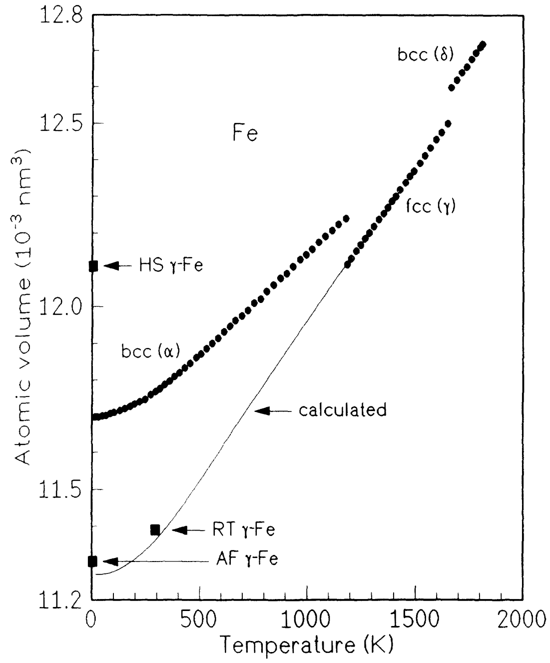

FIG. 3. The temperature dependence of the atomic volume of Fe. The AF, HS state and room-temperature (RT) values of the lattice constants, drawn with filled squares, are discussed in Sec. III. The calculated curve is obtained from the analysis in Sec. IV.

III. ATOMIC VOLUME OF γ-Fe

In Fig. 3, we show $V_a(T)$ of Fe.[23] In order to understand the behavior of γ-Fe outside its stability range, we look at the available experimental data on the atomic volume in fcc Fe alloys. The values of the room-temperature lattice constant for γ-Fe had been estimated previously from the extrapolation of the lattice constants of Fe-C and Fe-N alloys containing 2–8 at. % solute.[32] The atomic volume calculated from this extrapolated lattice constant 3.569 ± 0.005 Å is $(11.37\pm0.04)\times10^{-3}$ nm^3. This value is shown in Fig. 3 as RT γ-Fe. The lattice constant of γ-Fe determined on precipitates in Cu, $a=3.585$ Å, is not representative for equilibrium γ-Fe, since the precipitates reside under tension and adapt themselves to the larger Cu matrix, $a=3.615$ Å.

Using the room-temperature lattice constant and knowing the value at 1185 K, $a=3.647\pm0.001$ Å, we can now calculate an average thermal-expansion coefficient, α_{av}, between room temperature and the stability range. α_{av} is found to be $(24.5\pm1.9)\times10^{-6}$ K. Surprisingly, this value is larger than the measured value of the differential α of 23.3×10^{-6} K in the stability range, (cf. Fig. 1). Therefore, it follows that $\alpha(T)$ of γ-Fe must necessarily exhibit a maximum somewhere in the temperature interval 293 K$<T<$1185 K and be anomalously large, similar to that observed in the Fe-Ni-Mn alloys (cf. Fig. 2).

The analysis in Sec. IV requires the relative difference in volume between the AF ground and HS states at 0 K. We determine these values from the extrapolation of lattice constants at 4 K of different alloy series to γ-Fe. As shown in Fig. 4, to determine the lattice constant of the

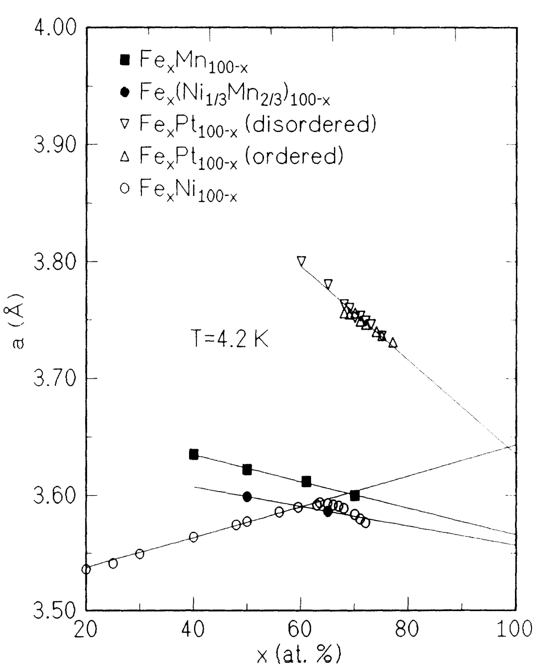

FIG. 4. The lattice constant of γ-Fe at 4 K in the AF and HS states. The AF state lattice constant is determined from the extrapolation of the lattice constants of the AF alloys Fe_xMn_{100-x} and $Fe_x(Ni_{1/3}Mn_{2/3})_{100-x}$. That for the HS state is determined from the FM alloys Fe-Ni, Fe-Pt, and Fe-Pd. Data for Fe-Pd are now shown for clarity.

AF state of γ-Fe AF fcc alloys, Fe_xMn_{100-x} and $Fe_x(Ni_{1/3}Mn_{1/3})_{100-x}$ are used (the latter alloy series has a constant electron concentration, $e/a=8$, equivalent to that of γ-Fe). For the HS state lattice constant, FM alloys of Fe-Ni, Fe-Pt, and Fe-Pd are used. From the figure, the lattice constant of γ-Fe in the AF state is determined to be 3.562±0.004 Å, which corresponds to a mean atomic volume of $(11.30\pm0.04)\times10^{-3}$ nm^3. The 4 K lattice constant of HS γ-Fe is 3.645±0.004 Å corresponding to an atomic volume of $(12.11\pm0.04)\times10^{-3}$ nm^3. These values are also shown in Fig. 3. The relative difference in the volume at 4 K between the two states is, consequently, 7%.

IV. EXCESS THERMAL EXPANSION IN γ-Fe

As mentioned previously, we use a thermal activation process to describe the enhancement of $\alpha(T)$ of γ-Fe, just as it was capable of describing the enhancement of $\alpha(T)$ in the fcc Fe alloys.

In a two-level system, denoted by LS and HS, the relative population at finite temperature $N(T)$ of the energetically higher level is given by

$$N(T)=\frac{1}{1+(g_{LS}/g_{HS})\exp(\Delta\varepsilon/kT)} , \qquad (1)$$

where g_{LS}/g_{HS} is the degeneracy ratio, k is the Boltzmann constant $\Delta\varepsilon$ is the energy difference between the two states, and T is the temperature. In the case of γ-Fe, LS≡AF. The volume per atom V is given by

$$V(T)=[1-N(T)]V_{LS}(T)+N(T)V_{HS}(T) , \qquad (2)$$

where V_{LS} and V_{HS} are the volumes per atom of the LS state and the HS state, respectively. The observed coefficient of thermal expansion $\alpha(T)$ is then

$$\alpha(T)=\frac{1}{3V}\frac{dV}{dT}\approx\frac{1}{3V_{LS}}\frac{dV_{LS}}{dT}+\frac{\Delta V}{3V_{LS}}\frac{dN}{dT}$$
$$=\alpha_{lat}^{LS}(T)+\alpha_{ex}(T) , \qquad (3)$$

where $\Delta V=V_{HS}-V_{LS}$. The first term in Eq. (3), $\alpha_{latt}^{LS}(T)$, is approximately the lattice thermal expansion of the pure LS state and the second term, $\alpha_{ex}(T)$, is an excess expansion. The first term is described by the Grüneisen relation,

$$\alpha_{lat}^{LS}(T)=\frac{c_v(T)}{3Q[1-KE(T)/Q]^2} , \qquad (4)$$

where c_V is the specific heat at constant volume, E is the internal energy, K is a constant associated with the indices of the Mie potential, and Q is a constant related to the Grüneisen constant γ through

$$\gamma=\frac{V}{\kappa Q} , \qquad (5)$$

where V is the atomic volume and κ is the compressibility. The second term in Eq. (3), with the aid of Eq. (1) can be written as

$$\alpha_{ex}(T)=\frac{\Delta V}{3V_{LS}}\frac{\Delta\varepsilon}{k^2T^2}\frac{(g_{LS}/g_{HS})\exp(\Delta\varepsilon/kT)}{[1+(g_{LS}/g_{HS})\exp(\Delta\varepsilon/kT)]^2} . \qquad (6)$$

Due to the enhanced nature of $\alpha(T)$ in the stability range of γ-Fe, $\alpha_{lat}^{LS}(T)$ cannot be estimated from Eq. (4) without obtaining unreasonable values for K and Q. This problem can be overcome by noting that the parameters involved in Eq. (6) can be estimated independently. Therefore, we first calculate $\alpha_{ex}(T)$ and then find an $\alpha_{lat}^{LS}(T)$ so that their sum will match the data in the stability range.

For the two parameters, g_{LS}/g_{HS} and $\Delta\varepsilon$, we use values previously determined for fcc Fe alloys from specific-heat measurements which also exhibit high-temperature enhancements.[15] The reason for this choice is that the sum of the lattice and electronic specific heats, which determines the "ground curve" of the measured data, are dependent on the Debye temperature Θ_D and the electronic γ coefficient. Since both of these parameters are determined from low-temperature measurements, the ground curve pertains essentially to the LS state. Any enhancement in the specific heat can, therefore, be unequivocally determined with respect to a pure LS state. This is not the case in a Grüneisen analysis for determining the lattice contribution to the thermal expansion since this analysis requires a fit to high temperatures which are already anomalous in the case of anti-Invar alloys. Values of g_{LS}/g_{HS} and $\Delta\varepsilon/k$ are plotted as a function of electron concentration in Fig. 5, from which we extrapolate for γ-Fe $(g_{LS}/g_{HS})\simeq0.74$ and $(\Delta\varepsilon/k)\simeq1350$ K.

$\alpha_{ex}(T)$ calculated from Eq. (6) with these data is shown in Fig. 6. Using $\Theta_D=430$ K for γ-Fe, $\alpha_{lat}^{LS}(T)$ can now be

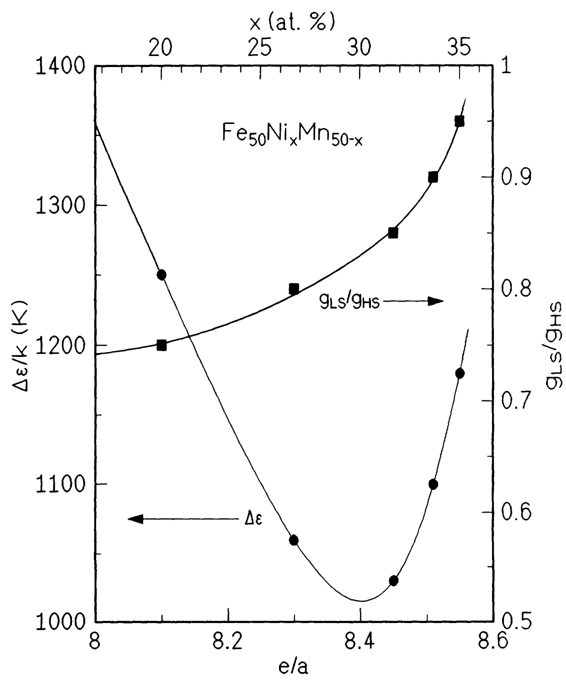

FIG. 5. $\Delta\varepsilon/k$ and g_{LS}/g_{HS} for the $Fe_{50}Ni_xMn_{50-x}$ alloy series. The values estimated for γ-Fe are 1350 and 0.74, respectively.

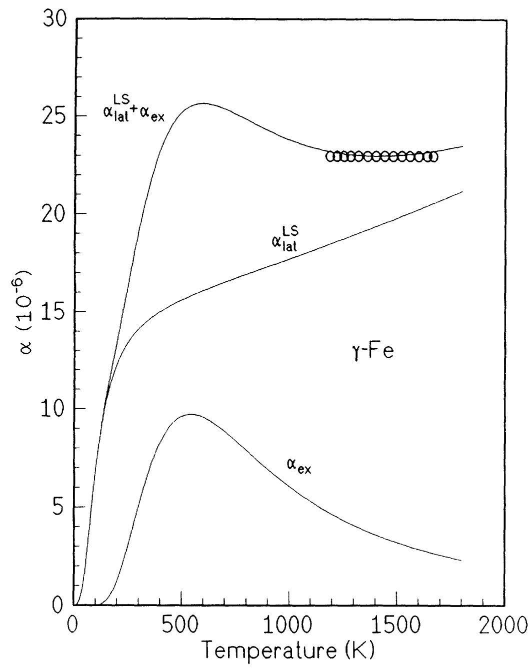

FIG. 6. $\alpha_{ex}(T)$, $\alpha_{lat}^{LS}(T)$, and $\alpha(T)=\alpha_{lat}^{LS}(T)+\alpha_{ex}(T)$ determined for γ-Fe.

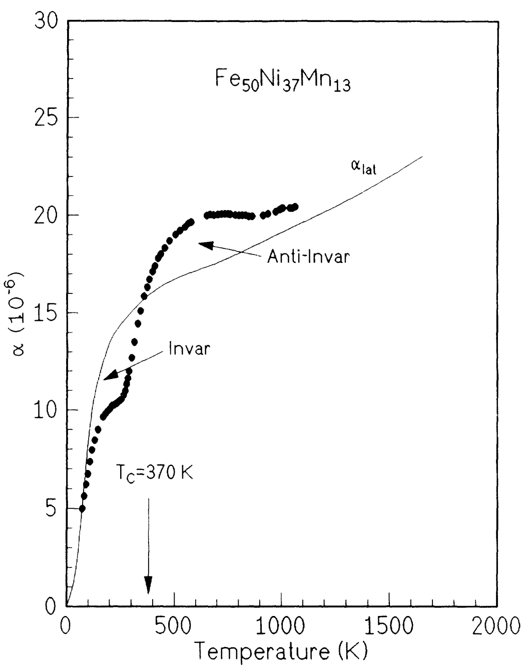

FIG. 7. $\alpha(T)$ of a FM ground-state $Fe_{50}Ni_{37}Mn_{13}$ sample with a Curie temperature $T_C=370$ K. This particular alloy exhibits the Invar effect below T_C and the anti-Invar effect above it.

determined by optimizing K and Q in Eq. (4) so that Eq. (3) gives the best fit to the experimental data in the stability range. With the values $K=2.1$ and $Q=550$ kJ/mole, $\alpha_{lat}^{LS}(T)$ results as shown in Fig. 6. The data clearly show that from the analysis given, a maximum in the thermal expansion of γ Fe results. This calls for the anti-Invar effect of γ-Fe.

Using the theoretical value[5] $\kappa=9.1\times10^{-12}$ m^2/N in Eq. (5), we obtain a Grüneisen constant, $\gamma=1.4$ for AF γ-Fe, supporting the results that the values of K and Q are reasonable. The volume enhancement determined from

$$\frac{\Delta V}{V}=3\int(\alpha-\alpha_{lat}^{LS})dT \tag{7}$$

amounts to about 2.8%. This shows that the anti-Invar effect in γ-Fe is larger than the Invar effect (2%) in $Fe_{65}Ni_{35}$.

$V_a(T)$ of γ-Fe in the complete temperature range, shown in Fig. 3, is calculated by integrating $\alpha(T)$. The calculated curves is seen to be in very good agreement with the values of the volumes estimated at 4 K and room temperature.

V. SUMMARY AND CONCLUSION

Using available data on γ-Fe inside the stability range and the values of the atomic volume at 4 K and room temperature, we have constructed $\alpha(T)$ of γ-Fe by using a two-level system model. The necessary parameters $\Delta\varepsilon$ and g_{LS}/g_{HS} have been adopted from specific-heat measurements on fcc Fe alloys.[15] $\alpha(T)$ of γ-Fe shows a maximum. The excess thermal expansion, $\alpha_{ex}(T)$, is the anti-Invar effect which leads to a volume enhancement of 2.8%. This is larger than the 2% spontaneous volume magnetostriction in $Fe_{65}Ni_{35}$ Invar. The calculated temperature dependence of the atomic volume of γ-Fe is in excellent agreement with the estimated values of the atomic volume at room temperature and at 4 K.

The enhancement in $\alpha(T)$ in γ-Fe is of the same order of magnitude as observed in the $Fe_{50}Ni_xMn_{100-x}$ alloys. As shown earlier, it exists in numerous other fcc alloys.[15] However, to determine absolute values of the volume enhancement in alloys with a reasonable degree of accuracy and to establish a general behavior as a function of electron concentration, as done for the Invar effect,[2,27] measurements extended to temperatures approaching the melting point are required.

As shown, the anti-Invar effect is a high-temperature property and a magnetic phenomenon occurring far above any magnetic ordering temperature. This is in contrast to the Invar effect which predominantly occurs below the respective magnetic ordering temperatures. Of specific interest are alloys such as $Fe_{50}Ni_{37}Mn_{13}$ which show, as demonstrated in Fig. 7, both an Invar effect in the magnetically ordered state and an anti-Invar effect above it. This interesting feature relating Invar and anti-Invar calls for further investigation.

ACKNOWLEDGMENT

The work was supported by the Deutscheforschungsgemeinschaft (SFB 166).

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