Low-temperature magnetization in Ni-rich γ -Ni_{100-x-v}Fe_xV_v alloys

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Detailed studies of the temperature dependence of the spontaneous magnetization have been made in eight different compositions of γ -Ni_{100-x-y}Fe_xV_y ($4 \le x \le 17$; $2 \le y \le 18$) alloys at a magnetic field of 30 kOe between 4.2 and 50 K. The values of the Curie temperature (T_C) and the saturation magnetization (M_0) are found to decrease with the increase of V. This is understood since the addition of V (which is antiferromagnetic) suppresses the ferromagnetic order in the NiFe binary alloys. The magnetization of the alloys with low V ($y \le 11$ at.%) has shown a good fit to only the spin-wave $T^{3/2}$ term whereas the data for the high V ($y \ge 12$ at.%) alloys require an additional Stoner T^2 term (for weak itinerant ferromagnets). This indicates that the addition of more V gives the alloys a weak itinerant electron character. The range of temperature of the above analysis is much lower than $0.1T_C$ for most of the alloys. Also, the Arrott plots near T_C show a set of almost parallel isothermal lines supporting the presence of the Stoner term. Thus the present magnetization studies show convincingly a transition from the localized to the weak itinerant electron ferromagnetism in the high V ($y \ge 12$ at.%) alloys.

I. INTRODUCTION

Magnetic properties of 3d transition-metal alloys have been a subject of great interest because of their unusual behavior. It is well known that any magnetic system is characterized by a magnetization which includes all relevant forms of magnetic interactions. Theoretically, it is not possible to consider all the magnetic interactions in 3d alloys to describe their magnetization. As a result, there is always some controversy in the interpretation of the experimental data. In the last few years, some binary and ternary alloy systems have attracted attention due to their complicated magnetic structures where both ferromagnetic and antiferromagnetic exchange interactions are found to compete with each other.²⁻⁴ Earlier, detailed neutron-diffraction studies^{2,3} on the ternary y-NiFeCr and y-NiFeV alloys showed that the exchange interactions between Ni-Ni is ferromagnetic; whereas, those for Fe-Fe, Cr-Cr, and V-V are antiferromagnetic. Interestingly, in Fe-rich γ-NiFeCr alloys, a host of different magnetic phases-ferromagnetic, spin glass, reentrant spin glass, and antiferromagnetic—have been observed within the same crystallographic fcc γ phase.^{4,5} As a result, most of the earlier work focused on those NiFeCr ternary alloys. Later, a detailed magnetization study revealed that the Ni-rich NiFeCr alloys become weak itinerant electron ferromagnets with increasing Cr content.⁶ In addition, a nonlinearity in the composition dependence of the magnetization has been recently observed, which is in contradiction with the split-band model.⁸ Other studies on amorphous as well as crystalline alloys indicated that the addition of Cr and Mn suppressed both the magnetic moment and the Curie tem-

perature (T_C) . 6,9-11 It is, in fact, very important to study the low-temperature magnetization of these alloys to understand better the role of different magnetic exchange interactions.⁵ NiFeV alloys have shown a change of sign in the extraordinary Hall (R_s) and the linear magnetostriction (λ_s) coefficients. The compositions, where $R_s = \lambda_s = 0$, are found to deviate strongly from the theoretically predicted line in the ternary phase diagrams. 12 This certainly indicates that there is some nonlinearity in the composition dependence of the magnetization.⁷ Another study predicted that addition of V might make the alloys weak itinerant electron ferromagnets.⁶ We have made detailed measurements of the temperature dependence of the dc magnetization in eight different compositions of γ -Ni_{100-x-v}Fe_xV_v ($4 \le x \le 17$; $2 \le y \le 18$) alloys at a magnetic field of 30 kOe in the temperature range of 4.2-50 K, which is well below their respective Curie temperatures. The composition of the present alloys varies in all the three constituents. Hence it will be very interesting to see how these composition variations affect the low-temperature magnetization. The present investigation is aimed at studying the low-temperature magnetization in terms of spin-wave and Stoner single-particle excitations to understand the process of thermal demagnetization. Also, it will be worthwhile to look for any signature of a transition to weak itinerant electron ferromagnetism with increasing V.

II. EXPERIMENTAL DETAILS

The alloys were prepared by induction melting of "specpure" (5N purity) grade constituent elements, obtained from Johnson-Mathey, Inc. (England). For homogenization, the al-

| TABLE I. Alloy compositions along with their sample designation, Curie temperature (T_C) , density, values of saturation magnetization |
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| (M_0) , equations of fit, fitting parameters, values of χ^2 , and stiffness constant D_0 . |

| Sample designation | Alloy composition Ni-Fe-V (at. %) | T_C (K) | Density ρ (g/cm ³) | M_0 (emu/g) | Equation of fit for $[\Delta M(T)/M(0)]$ | B (10 ⁻⁵ K ^{-3/2}) | $\alpha (10^{-5} \mathrm{K}^{-2})$ | χ^2 (10 ⁻⁹) | D_0 (meV Å ²) |
|--------------------|---|--------------|-------------------------------------|---------------|--|---|------------------------------------|------------------------------|-----------------------------|
| 1 | 81-17-2 | 746±3 | 8.67 | 55.2 | $-BT^{3/2}Z(3/2,T_g/T)$ | 2.2±0.1 | | 4.3 | 190 |
| 2 | 83-10-7 | 486 ± 2 | 8.60 | 56.7 | -do- | 2.1 ± 0.1 | | 2.0 | 193 |
| 3 | 80.5-10.5-9 | 417 ± 3 | 8.54 | 64.5 | -do- | 1.0 ± 0.1 | | 0.7 | 288 |
| 4 | 77-12-11 | 393 ± 1 | 8.45 | 47.2 | -do- | 3.2 ± 0.1 | | 1.6 | 167 |
| 5 | 82.5-5.5-12 | 362 ± 3 | 8.54 | 41.2 | $-BT^{3/2}Z(3/2,T_g/T)-\alpha T^2$ | 1.0 ± 0.2 | 0.5 ± 0.06 | 2.6 | 394 |
| 6 | 77-7-16 | 155 ± 1 | 8.38 | 26.7 | -do- | 15.0 ± 3.0 | 2.0 ± 0.6 | 11.2 | 88 |
| 7 | 79-5-16 | 62 ± 0.5 | 8.40 | 16.2 | -do- | 59.0 ± 4.0 | 6.0 ± 0.8 | 9.3 | 49 |
| 8 | 78-4-18 | 43 ± 0.5 | 8.35 | 13.9 | | | | | |

loy ingots were annealed at 1100 °C for 48 h under argon atmosphere and then quenched to room temperature (in water) to keep their high-temperature crystallographic fcc γ-phase as well as their random substitutional disorder.⁶ The crystallographic phase and the alloy homogeneity were checked using the powder x-ray-diffraction technique and the energy dispersive x-ray analysis, respectively. The dc magnetization was measured with Lake-shore Model 7229 Extraction Magnetometer/Susceptometer. The samples were cut in a cylindrical shape where the length/diameter ratio is large so that for magnetic fields, applied along the length of the sample, the demagnetization factor (N) is negligibly small $(N \cong 10^{-3})$. The measurements have been done at every 0.05 K in the temperature range of 4.2-15 K and in steps of 0.1-0.3 K up to 50 K at a magnetic field of 30 kOe. The resolution in the magnetization data is found to be of the order of 1 in 10^5 .

III. RESULTS AND DISCUSSION

The alloy composition along with their sample designation, the values of \boldsymbol{T}_{C} , and the saturation magnetization \boldsymbol{M}_{0} (extrapolated to T=0 K) are given in Table I. Like Ni-rich NiFeCr alloys, an other low-temperature magnetic phases (e.g., spin glass, reentrant spin glass, and antiferromagnet) were observed in the present alloys down to 4.2 K. For convenience, from now on we will refer to the alloys by their respective sample designation. For the low V ($y \le 12$) alloys, the values of T_C are taken from an earlier work, whereas, those for the high V ($y \ge 16$) are taken from our recent study. 13 It is observed from Fig. 1 that the value of M_0 remains almost constant up to V concentration of 11 at. %, beyond which there is a sharp fall. On the other hand, T_C (Fig. 1) decreases almost linearly with the increase in V. The increase in V in these bulk alloys enhances the contribution of the antiferromagnetic exchange interaction ¹⁴ for V-V and Fe-V pairs which, in turn, suppresses the ferromagnetic order in the alloys resulting in lower M_0 and T_C . The same antiferromagnetic pair interaction beyond a certain critical concentration of V is responsible for the decrease in M_0 in V-rich $(y \ge 12)$ alloys. The recent ac-susceptibility measurements¹³ do show only ferromagnetic phase and no other (say, spin-glass-like) phase at low temperatures in the present set of bulk NiFeV alloys. Nevertheless, the above behavior of M_0 and T_C is similar to the earlier results on crystalline and amorphous alloys^{6,11} containing Cr and Mn.

At very low temperatures well below T_C , the temperature dependence of the spontaneous magnetization of any 3d ferromagnetic material is generally described by spin-wave (SW) theory. The dispersion relation for spin waves, in the long-wavelength limit, is given by

$$\varepsilon(q) = g \mu_B H_{\text{int}} + Dq^2 + Eq^4 + \dots \tag{1}$$

where ε is the energy of spin-wave excitations and q is the wave vector. The first term $(g \mu_B H_{\text{int}} \ll Dq^2)$ of Eq. (1) represents an energy gap in the presence of an effective internal field H_{int} . The internal field H_{int} is defined as

$$H_{\rm int} = H_{\rm ext} - NM_0, \tag{2}$$

where $H_{\rm ext}$ is the externally applied magnetic field and N is the demagnetization factor. In Eq. (1), D is the spin-wave stiffness constant and E is a proportionality constant. In the low-temperature limit, according to the Heisenberg model, the change in the spontaneous magnetization due to spin-wave excitations 15,16 can be written as

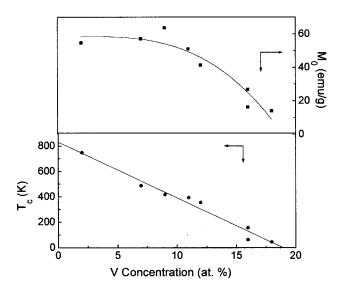


FIG. 1. Dependence of the saturation magnetization (M_0) and the Curie temperature (T_C) on the V concentration in NiFeV alloys. The lines are guides to the eye.

$$\Delta M(T)/M(0) = [M(T) - M(0)]/M(0)$$

$$= -B_1 Z(\frac{3}{2}, T_g/T) T^{3/2} - C_1 Z(\frac{5}{2}, T_g/T) T^{5/2},$$
(3)

where $T_g = g \mu_B H_{\rm int}/k_B$ is the gap temperature, and $Z(\frac{3}{2},T_g/T)$ and $Z(\frac{5}{2},T_g/T)$ are two standard correction terms. The coefficients B_1 and C_1 ($B_1,C_1>0$) in Eq. (3) are related to the spin-wave stiffness constant D and the average mean-square range $\langle r^2 \rangle$ of the exchange interaction. In Eq. (3), the $T^{3/2}$ term comes from the harmonic term (q^2) of the spin-wave dispersion relation whereas $T^{5/2}$ term originates from the anharmonic one (q^4). In the above relations, the spin-wave stiffness constant D is considered to be temperature independent. But, theoretically, it has been found that the interaction between spin waves and itinerant electrons leads to a temperature dependence of $D \propto T^2$; The whereas, magnon-magnon interactions can give rise to a $D \propto T^{5/2}$ dependence. Hence, in the low-temperature limit, the spin-wave stiffness constant D can be written as

$$D = D_0(1 - D_1 T^2 - D_2 T^{5/2}). (4)$$

Substitution of Eq. (4) into Eq. (3) gives

$$[\Delta M(T)/M(0)]_{sw} = -BT^{3/2}(1 - D_1T^2 - D_2T^{5/2})^{-3/2} \times Z(3/2, T_g/T) - CT^{5/2}Z(5/2, T_g/T),$$
(5)

where $B=B_1(D=D_0)$ and C(B,C>0) are two constants. In addition to the $T^{3/2}$ and $T^{5/2}$ terms, the binomial expansion of the first term of Eq. (5) gives rise to additional temperature dependences of $T^{7/2}$ and T^4 .

Besides the spin-wave excitations, the Stoner singleparticle excitations also contribute to the low-temperature demagnetization of a ferromagnet. ^{21,22} The basic idea behind this model is that the magnetic electrons are split into spin-up and spin-down subbands which are separated by the exchange energy. In other words, the energy difference between the split bands is directly proportional to the spontaneous magnetization. In the very low-temperature limit, thermal excitation of itinerant electrons from the majority to the minority band (with increasing temperature) gives rise to a decrease in the magnetization. Later on, it was realized that the temperature dependence of magnetization also depends on whether the contributions to magnetization are coming from electrons (or holes) of a single subband or both subbands.²³ The general expression for the reduced magnetization due to Stoner excitations is given by

$$[\Delta M(T)/M]_{\rm sp} = -AT^n \exp(-\Delta/k_B T), \tag{6}$$

where Δ is the energy gap between the top of the full subband and the Fermi level E_F , and the proportionality constant $A\!>\!0$ depends on the band parameter. In strong ferromagnets (e.g., Ni and Co where one subband is completely full, i.e., $\Delta\!\neq\!0$), the change in magnetization comes from the single-particle excitations of one partially filled subband and is given by Eq. (6) with $n\!=\!\frac{3}{2}$.

On the other hand, in weak ferromagnets [e.g., Fe where both the subbands are partially filled ($\Delta \approx 0$) and n=2], the reduced magnetization can be expressed as

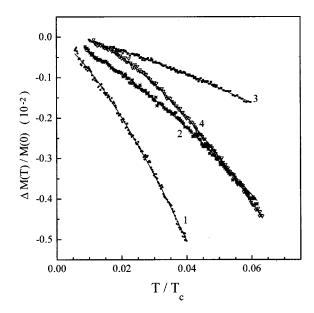


FIG. 2. Change in reduced magnetization $[\Delta M(T)/M(0)]$ as a function of reduced temperature (T/T_C) for alloys 1 $(Ni_{81}Fe_{17}V_2)$, 2 $(Ni_{83}Fe_{10}V_7)$, 3 $(Ni_{80.5}Fe_{10.5}V_9)$, and 4 $(Ni_{77}Fe_{12}V_{11})$. The solid lines are fits of the experimental data to Eq. (10) without the Stoner term.

$$[\Delta M(T)/M]_{\rm sp} = -\alpha T^2, \tag{7}$$

where $\alpha > 0$ is the proportionality constant related to various band parameters, like the density of states at the Fermi level, band splitting, etc. At very low temperatures $(T \leq T_C)$, the excitations from the spin-wave and the single-particle excitations are nearly independent and hence the final expression for the thermal demagnetization can be written as

$$\Delta M(T)/M = [\Delta M(T)/M]_{sw} + [\Delta M(T)/M]_{sp}.$$
 (8)

The temperature dependence of the reduced magnetization $\lceil \Delta M(T)/M(0) \rceil$ is shown in Figs. 2 and 3 for all the alloys

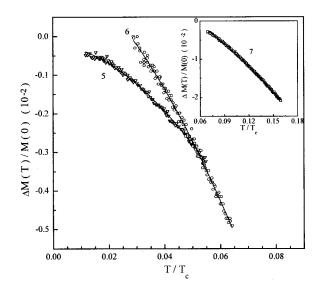


FIG. 3. Change in reduced magnetization $[\Delta M(T)/M(0)]$ as a function of reduced temperature (T/T_C) for alloys 5 $(Ni_{82.5}Fe_{5.5}V_{12})$, 6 $(Ni_{77}Fe_7V_{16})$, and 7 $(Ni_{79}Fe_5V_{14})$ (inset). The solid lines are fits of the experimental data to Eq. (10).

except 8. It is very important to note here that the present temperature range is much below their respective 0.1 T_C (except for samples 7 and 8). This makes the interpretation of the data meaningful in terms of the spin-wave and Stoner single-particle low-temperature excitations. However, for alloy 8 ($T_C \cong 43 \text{ K}$), the data could not be meaningfully analyzed in the temperature range of 4.2-16 K. In general, we have tried to fit the data first to only the spin-wave excitations [Eq. (5)]. The gap temperature $T_g = g \mu_B H_{\text{int}}/k_B$ in the correction terms of $Z(\frac{3}{2}, T_g/T)$ and $Z(\frac{5}{2}, T_g/T)$ can be estimated if $H_{\rm int}$ is known [see Eq. (2)]. As mentioned earlier, the demagnetization factor in the present samples is found to be quite small $(N \approx 10^{-3})$. Thus $H_{\rm int} \cong H_{\rm ext}$. The calculated value of T_g , using g = 2 and $H_{\text{int}} = 30$ kOe, is found to be 4.0 K and this has been incorporated as a constant in the fitting program. The fitting of the data shows that only the $T^{3/2}$ term gives a considerably good fit for alloys 1 to 7. Inclusion of higher-order terms like $T^{5/2}$ and/or $T^{7/2}/T^4$, in addition to the $T^{3/2}$ [see Eq. (5)], yields a negative coefficient for the higherorder terms which is unphysical. Similar results have been found earlier in Ni-rich crystalline NiFeCr 6,14 and Co-rich amorphous alloys. ¹¹ They show that the $T^{5/2}$ and $T^{7/2}/T^4$ terms of the spin-wave excitations have negligible contributions to the temperature dependence of magnetization. This could very well be true in the present NiFeV alloys as well. However, we could not make any estimate for the contributions of the $T^{5/2}$ and $T^{7/2}/T^4$ terms from our data. On the contrary, it is found necessary to consider the Stoner singleparticle excitations, besides the spin-wave term, to describe better the temperature dependence of the magnetization convincingly. Hence the equations for fitting are modified as follows:

$$\Delta M(T)/M(0) = [\Delta M(T)/M]_{\text{sw}} + [\Delta M(T)/M]_{\text{sp}}$$

$$= -BT^{3/2}Z(\frac{3}{2}, T_g/T) - \beta T^{3/2} \exp(-\Delta/k_B T)$$
(for strong ferromagnets) (9)
$$= -BT^{3/2}Z(\frac{3}{2}, T_g/T) - \alpha T^2$$
(for weak ferromagnets). (10)

A fit of the magnetization data to Eq. (9) gives unphysical values of the fitting parameters β and Δ for all the alloys. This is not unexpected since too many parameters generally make a fit unphysical. Moreover, a recent study²⁴ on some Ni-rich NiFeCr alloys has shown that the 3d band splitting of Ni, Fe, and Cr or V are not so explicit (Δ is very small) as is envisaged in the virtual bound state or the Stoner models. Thus strong itinerant electron ferromagnetism is ruled out in these NiFeV alloys. On the other hand, fitting to Eq. (10) shows that the data for the high V ($y \ge 12$ at. %) alloys (i.e., samples 5, 6, and 7 in Table I) give a good fit with normalized $\chi^2 = 1/(N-1)\{\Sigma[Y_i(\text{fit}) - Y_i(\text{data})]^2/[Y(\text{mean})]^2\}$ of the order of $(1-10)\times10^{-9}$ which is consistent with the present experimental resolution (1 in 10⁵)). However, for the low V ($y \le 11$ at. %) alloys, a fit to Eq. (10) gives a negative value for B, which is unphysical. This indicates that the data for the low V alloys can be described by the spin-wave $T^{3/2}$ term only. All the fitting parameters are given in Table I. The

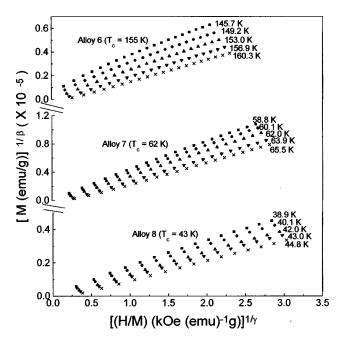


FIG. 4. Arrott plots $[M^{1/\beta} \text{ vs } (H/M)^{1/\gamma} \text{ isotherms}]$ for alloys 6 $(\text{Ni}_{77}\text{Fe}_7\text{V}_{16})$, 7 $(\text{Ni}_{79}\text{Fe}_5\text{V}_{14})$, and 8 $(\text{Ni}_{78}\text{Fe}_4\text{V}_{18})$ at several temperatures near their T_C 's.

best-fitted curves along with the raw data are shown in Figs. 2 and 3 for the low and the high V alloys, respectively. The fits are indeed good, keeping in mind the fact that the change of magnetization is even less than 0.5% in this lowtemperature range. The values of B are found to increase as the V concentration increases. This is in good agreement with the earlier studies on NiFeCr alloys. The values of the spin-wave stiffness constant D_0 , calculated from the coefficient $B = [2.612\mu_B/M(0)\rho][k_B/4\pi D_0]^{3/2}$ where ρ is the density of the alloy}, decreases as the V concentration increases beyond 12 at. % (see Table I). A similar decrease in D_0 with increasing Cr concentration was reported earlier. 6,11,14 However, the values of α are found to be two orders of magnitude larger than those observed in NiFeCr alloys. This is quite puzzling. Nevertheless, the present study clearly shows that, as the V concentration increases, the alloys behave more like weak itinerant ferromagnets.

Moreover, it is interesting to look for other signatures for the weak itinerant electron character in these NiFeV alloys. In itinerant ferromagnets, the Arrott plots are found to be a set of straight lines at high fields over a wide temperature range near T_C . ²⁵ This kind of relationship arises from the Landau theory of phase transitions. ²⁶ The slopes of such lines, however, are temperature dependent. Later, Edward and Wohlfarth ²⁷ demonstrated that in the limit of weak itinerant ferromagnetism the expression for the magnetization is given by

$$\left[\frac{M(H,T)}{M(0,0)}\right]^{1/\beta} = \left[1 - \left(\frac{T}{T_C}\right)^2\right] + \left[\frac{2X_0H}{M(H,T)}\right]^{1/\gamma}, \quad (11)$$

where β and γ are the critical exponents for paramagnetic-to-ferromagnetic phase transitions, and X_0 is a proportionality constant. This shows that the Arrott plots at all temperatures near T_C would be a set of temperature-independent parallel lines with a slope equal to $[2X_0M(0,0)^{1/\beta}]^{1/\gamma}$. In one

of the recent studies, ¹³ the values of the critical exponents β and γ are found to be as 0.25, 0.21, and 0.22, and 1.01, 0.99, and 1.01 for alloys 6, 7, and 8, respectively. In Fig. 4, the Arrott plots for alloys 6, 7, and 8 are shown at the temperatures very near to their respective T_C 's. We find that the plots are a set of nearly parallel lines for all the alloys. The latter is an important finding in the present investigation and clearly supports our conclusion that the alloys with high V ($y \ge 12$ at. %) become weak itinerant ferromagnets. To sum up, this is the only study on γ -NiFeV alloys where the temperature dependence of spontaneous magnetization has been convincingly interpreted. It is also shown that the addition of V gives the NiFeV alloy a weak itinerant electron character.

IV. CONCLUSIONS

To conclude, detailed studies of the temperature dependence of the dc magnetization are presented for eight different compositions of γ -Ni_{100-x-y}Fe_xV_y (4 \leq x \leq 17; 2 \leq y \leq 18) alloys at a magnetic field of 30 kOe between 4.2 and 50 K. The values of the saturation magnetization (M_0) and the Curie temperature (T_C) are found to decrease as the V content increases. This is expected since the increase in V (where the V-V and Fe-V exchange interactions are antiferromagnetic) suppresses the ferromagnetic ordering in the al-

loys. The magnetization of the alloys with low V $(y \le 11 \text{ at. \%})$ have shown a good fit to only the spin-wave $T^{3/2}$ term. On the other hand, the data for the high V $(y \ge 12 \text{ at. \%})$ alloys are found to fit to the sum of the spin-wave $T^{3/2}$ and the Stoner T^2 terms. This indicates that the present alloys with high V content $(y \ge 12 \text{ at. \%})$ tend to become weak itinerant ferromagnets. The range of temperature of this analysis is much lower than $0.1\,T_C$ of the respective alloys (except for the alloys 7 and 8). This is very important for a meaningful interpretation of the data in terms of spin-wave and Stoner single-particle excitations. The Arrott plots near T_C show a set of almost temperature-independent parallel lines which supports the above findings. This is clearly a manifestation of a transition to weak itinerant electron ferromagnetism in the high V $(y \ge 12 \text{ at. \%})$ alloys.

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