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Spin-wave dispersion relations in disordered Fe-V alloys

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The spin-wave dispersion relations of the ferromagnetic disordered alloys $Fe_{1-x}V_x$ ($x = 0.076$, 0.135, 0.160, and 0.187) were studied by means of the inelastic scattering of neutrons. The observed dispersion relations are adequately represented by the power law, $E = Dq^2(1-\beta q^2)$, in a wide energy range up to 80 meV. The concentration dependence of the exchange stiffness constant D shows good agreement with previous results obtained by means of the small-angle scattering of neutrons and by the analysis of the temperature dependence of the bulk magnetization. The observed results can be explained by the Heisenberg model and, to some extent, by the itinerant-electron model.

I. INTRODUCTION

The spin-wave dispersion relations of ferromagnetic metals and alloys with parallel moments are expressed in most cases by the power law,

$$E = Dq^2(1-\beta q^2) \ , \qquad (1)$$

for small values of q, where E, q, and D are the spin-wave energy, spin-wave momentum, and stiffness constant, respectively. For example, for Ni-based ferromagnetic alloys in which the magnetic moments of Ni and the impurity atoms are parallel, the experimentally determined spin-wave energy can be described by the above power law.[1-3] However, if the orientation of the magnetic moment of the impurity atoms with respect to the host moment is taken into consideration, deviations from the power law are predicted theoretically. Several theories show the existence of extra spin-wave modes in a Heisenberg ferromagnet containing an isolated impurity which has its magnetic moment antiparallel to those of host atoms.[4-7] The experimental evidence for the resonance modelike anomaly in a powder sample of Fe-Mn was reported.[8] Fe-V alloys are considered to be one of the most appropriate systems for studying the deviation from the power law, because V atoms have moments of about $1\mu_B$ aligned antiparallel to the host moment of $2.2\mu_B$ and because single-crystal specimens can be easily grown.[9-11]

The exchange stiffness constants of several Fe-V alloys were determined previously by means of the small-angle scattering of neutrons[12] and by the analysis of the temperature dependence of the bulk magnetization.[13] In the former experiment, the stiffness constant was determined from the spin waves with small momentum, assuming the q^2 dependence of the spin-wave energy [corresponding to $\beta = 0$ in Eq. (1)]. In the latter case, it was estimated from the average density of states for the spin waves on the assumption that their energies can be expressed by the power law of Eq. (1)

A direct determination of the stiffness constant can be made by the inelastic neutron scattering experiment on a single crystal using a triple-axis spectrometer. In the present study, the spin-wave dispersion relations of Fe-V alloys with the V concentration from 7.6 to 18.7 at.% were measured for energies ranging from 5 to 80 meV. The purpose of the measurement was to observe possible anomalies in the dispersion relation as well as to determine the stiffness constants D and β for these alloys.

II. EXPERIMENTS

Four specimens containing 7.6, 13.5, 16.0, and 18.7 at.% V were grown into single crystals of about $0.2-1.0\ cm^3$ in volume by melting raw materials with 99.9% purity in a zirconia crucible placed in a Bridgman furnace. The specimens containing 7.6 and 13.5 at.% V were annealed at about 1000 °C for a day and then water-quenched. The 16.0 and 18.7 at.% V alloys received no heat treatment.

Neutron scattering experiments for the 16.0 at.% V specimen were performed by using the TUNS triple-axis spectrometer at the JRR-2 in Japan Atomic Energy Research Institute, and the HB2 and the HB3 triple-axis spectrometers at the HFIR in the Oak Ridge National Laboratory were used for the measurements on the other specimens. The spin-wave peaks were observed at room temperature by the constant-energy-transfer technique. The (002) reflection of pyrolytic graphite was used as the monochromator and the analyzer for the TUNS. The (101) reflection of Be was chosen as the monochromator and the analyzer for the HB2 spectrometer. Be and pyrolytic graphite were used as the monochromator and the analyzer for the HB3 spectrometer, respectively. The spin-wave peaks were observed along the $\langle 001 \rangle$ and the $\langle 110 \rangle$ directions around the 110 reciprocal-lattice point, but no anisotropy of the dispersion relation was detected. As a reference, the spin-wave peaks of pure Fe were also measured with the same experimental conditions.

Because of the very steep dispersion relation for the alloys the effect of the finite instrumental resolution should be corrected carefully, even though the spin-wave dispersion relation of pure Fe determined with the same experimental condition agrees, without any resolution correction, with the previous results obtained by Mook *et al.*[14] and Shirane *et al.*[15] The instrumental parameters, such as the mosaicities of the monochromator and the analyzer crystals and the angular divergences of the collimators, were chosen so that the observed line profiles of the Bragg reflections could be reproduced. Some typical results of constant E scans are shown in Fig. 1 together with the calculated intensity distributions which include the effects of the instrumental resolution. Arrows in the figure indicate the positions of the momentum corresponding to the spin-wave energies. It can be seen that small but significant shifts of the peak positions exist for the spin waves in the Fe-V alloys due to resolution effects.

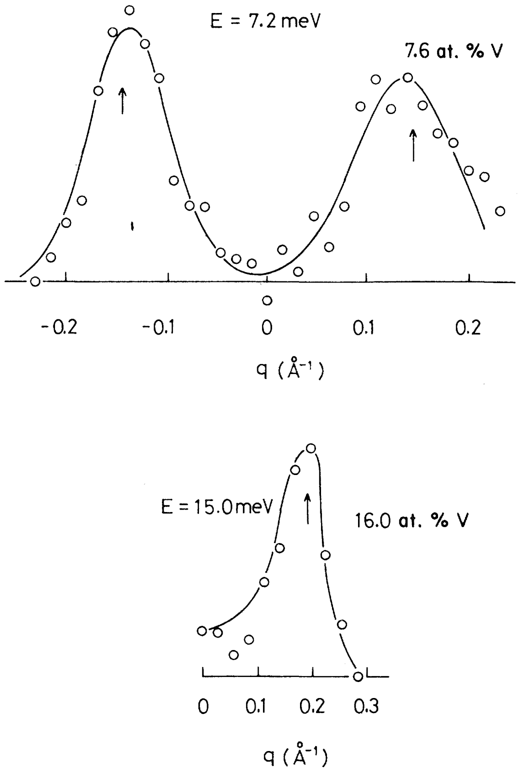

FIG. 1. Examples of scattered neutron groups. Solid lines are the intensities calculated by convoluting the dispersion curve given by Dq^2 with the resolution function. Arrows indicate the position of the true spin-wave momentum transfer corresponding to the neutron energy transfer.

III. RESULTS

The spin-wave dispersion relation for each specimen at room temperature was determined as shown in Fig. 2 after the resolution function corrections were made. The smallest momentum transfer observable in the present experiment is restricted by the interference due to high intensity phonon peaks. Broken curves in Fig. 2 show the dispersion curves evaluated by using the stiffness constants deduced from the results of Lowde *et al.*[12]

The spin-wave energy E is usually expressed in terms of the power of the momentum q as shown by Eq. (1). This equation, though neglecting the terms higher order than q^4, represents fairly well the experimentally determined spin-wave dispersion relations of Fe,[14,15] Ni,[16] and other alloys.[1-3] If the experimental results can be represented by Eq. (1), E/q^2 plotted against q^2 should be exactly a straight line. Such a plot was made for each spectrum as shown in Fig. 3 and no significant deviation from a straight line was found in the energy range between 5 and 80 meV, indicating that there is no resonancelike anomaly of the spin waves in these alloys. The stiffness constant D and the parameter β were determined from these plots and the numerical values are given in Fig. 3. The dispersion relations calculated from these D and β are shown in Fig. 2 by solid curves. The values of D and β for the alloys are shown in Fig. 4 together with those for pure Fe obtained by Mook *et al.*,[14] Shirane *et al.*,[15] and the present experiment. The squares and triangles in Fig. 4 are the values of D at room temperature for Fe-V alloys which we calculated from the values at absolute zero temperature obtained by Lowde *et al.*[12] and by Aldred,[13] respectively, by using the conversion factor determined by Aldred. These results obtained by different methods agree well with each other.

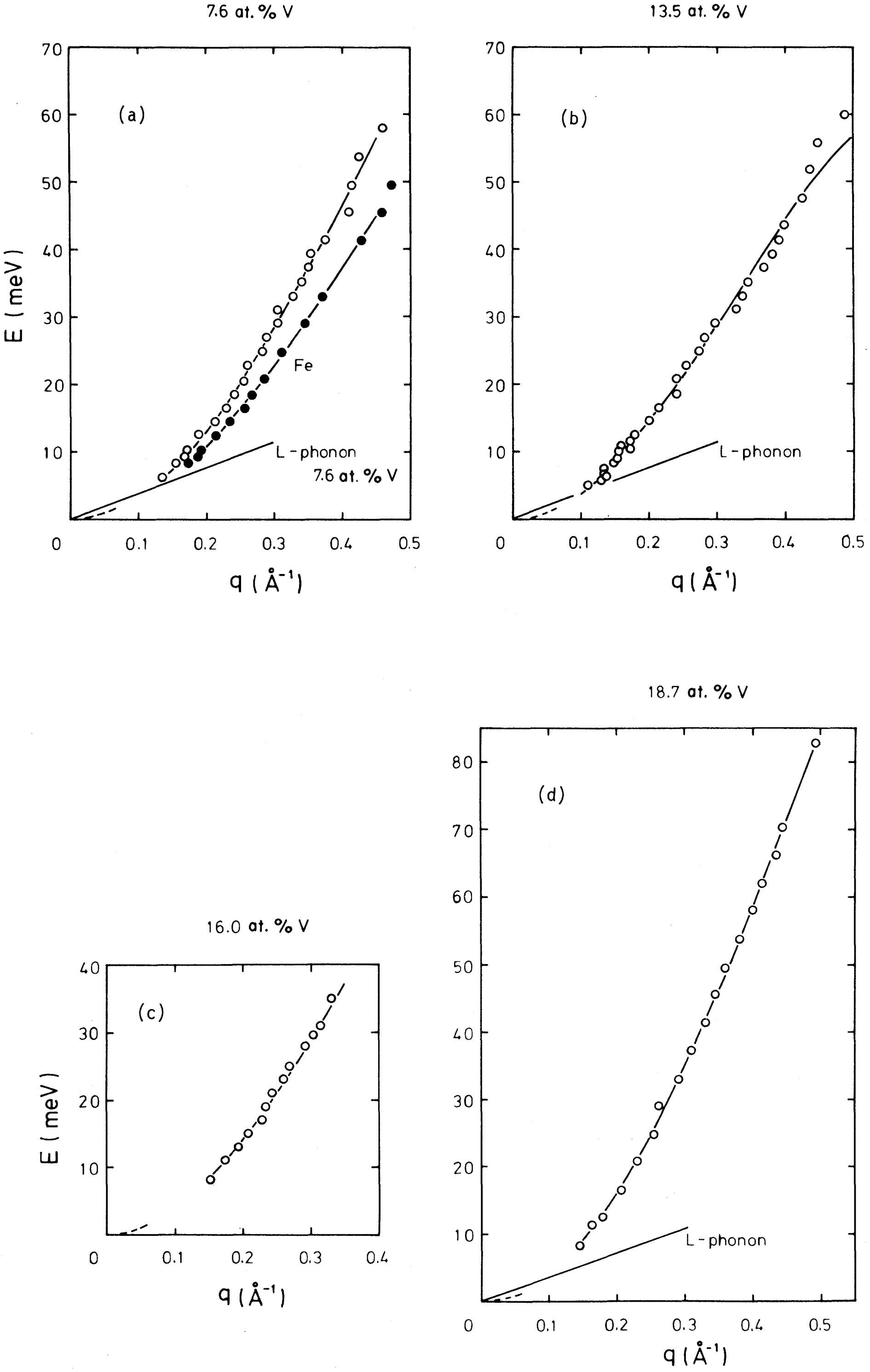

FIG. 2. Spin-wave dispersion curves for Fe-V alloys containing (a) 7.6 at.% V and pure Fe, (b) 13.5 at.% V, (c) 16.0 at.% V, and (d) 18.7 at.% V. Solid curves show the calculated dispersion relation given by Eq. (1) with the stiffness constant and the parameter β determined by the present experiment. Broken lines near the origin represent the dispersion relation given by D deduced from the work of Ref. 12.

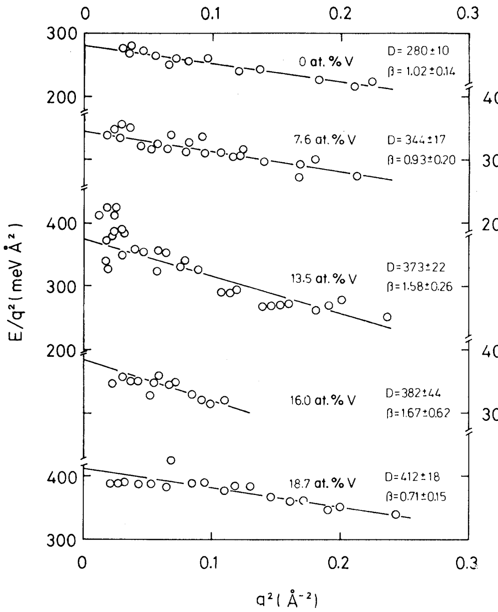

FIG. 3. E/q^2 as a function of q^2 for each specimen. Linearly extrapolated values of E/q^2 to $q^2=0$ and the slopes of the best-fitted straight lines give the stiffness constants D and the parameters β.

IV. DISCUSSION

As seen in Fig. 4, the exchange stiffness constants D estimated from the data in the region of $q > 0.25$ Å^{-1} and $E > 5$ meV agree fairly well with the values obtained at $q \lesssim 0.05$ Å^{-1} by Lowde *et al.*[12] using the small-angle scattering technique. It is concluded, therefore, that Eq. (1) adequately describes the spin-wave spectrum of the Fe-V alloys in a wide range of the excitation energy, as it does for other metals and alloys. These results may be interpreted on the basis of two different pictures, the Heisenberg model, and the band model.

First we consider the analysis based on the Heisenberg model for the virtual metal corresponding to the alloy $Fe_{1-x}V_x$. The spin-quantum number of the virtual atom, $\bar{S}$, which is defined as $(1-x)S_{Fe}+xS_V$, can be deduced from the bulk magnetization data. S_V can be determined from a neutron measurement of the magnetic moment on V in the alloy.[11] It is assumed that the exchange constant between the virtual atoms, $\bar{J}$, and that between the virtual atom and V, J', can be written as

$$\bar{J}\bar{S}^2 = (1-x)^2 J_{Fe-Fe} S_{Fe} S_{Fe} + 2x(1-x) J_{Fe-V} S_{Fe} S_V + x^2 J_{V-V} S_V S_V ,$$
$$J'\bar{S}S_V = (1-x) J_{Fe-V} S_{Fe} S_V + x J_{V-V} S_V S_V , \tag{2}$$

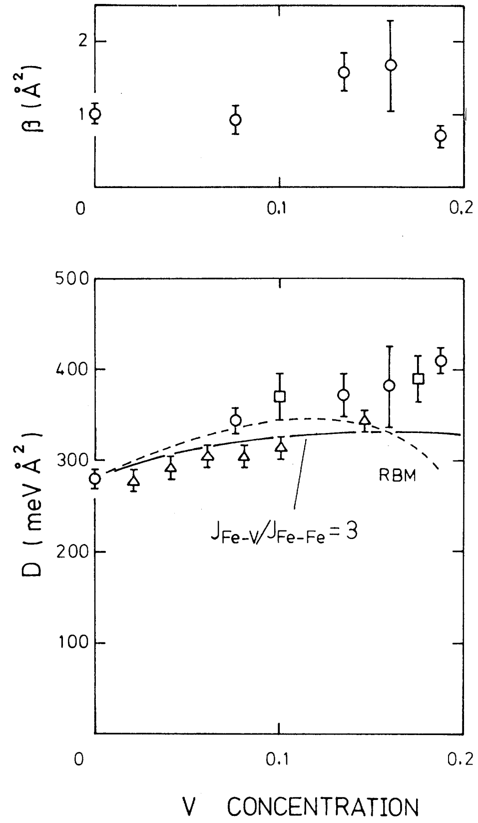

FIG 4. Concentration dependence of the stiffness constant and the parameter β. Squares and triangles are the values at room temperature calculated from the values at absolute zero temperature obtained by Lowde *et al.* (Ref. 12) and Aldred (Ref. 13), respectively. The conversion factor determined by Aldred was used. The solid and broken curves display the stiffness constants calculated in the nearest-neighbor Heisenberg model and in the rigid-band itinerant-electron model (RBM), respectively.

where $\bar{J}_{i-j}$ means the exchange parameter between the ith and jth species. The terms containing J_{V-V} are neglected. Extending the expression given by Child and Cable[10] for the concentration dependence of the Curie temperature T_C, in low concentration alloys, one can write the concentration dependence for the virtual metal as

$$\frac{1}{T_C}\frac{dT_C}{dx}\bigg|_{x=x_0} = \left[\frac{J'(x_0)S_V}{\bar{J}(x_0)\bar{S}(x_0)}\right]^2 - 1 . \tag{3}$$

From Eqs. (2) and (3), together with dT_C/dx estimated from the values of T_C given in Ref. 17, the ratio J_{Fe-V}/J_{Fe-Fe} was found to be approximately equal to 3 in the concentration range for the alloys of the present study. Using the value J_{Fe-Fe} which is consistent with the value of the stiffness constant for pure Fe, one can calculate the stiffness constant D for the alloy within the molecular-field approximation by the expression

$$D=(z/3)R_n^2\overline{JS} \ , \tag{4}$$

where R_n and z are the nearest-neighbor distance and the coordination number, respectively. The result of the calculation is shown by the solid line in Fig. 4.

In the simple band picture of ferromagnetism involving only one exchange splitting constant, D is given as[18]

$$D=\frac{1}{6\bar{S}\Delta}\left|\frac{\Delta}{2}[M(E_+)+M(E_-)]-\int_{E_-}^{E_+}M(E)\,dE\right| \ , \tag{5}$$

where

$$M(E)=\frac{V_0}{8\pi}\sum_{\lambda}\int_{E_{\lambda k}=E}|\nabla E_\lambda|\,dS \ ,$$

and $\Delta=E_+-E_-$. $E_+(E_-)$ is the Fermi energy for the electrons with up (down) spin, V_0 is the volume of the crystal, and λ is the band index. The function $M(E)$ and the density of states $N(E)$ for bcc Fe are given by Wakoh,[18] but the D for Fe calculated from Eq. (5) was found to be too small by factor of 3.9 in comparison with the experimental value. Nevertheless, it may be of some interest to calculate the x dependence of D in this approximation. The band splitting, Δ, for each alloy was estimated from $N(E)$ and the average magnetic moment $2\bar{S}$. The calculated value of D was then multiplied by 3.9 to reproduce the correct value for Fe. The resulting x dependent D is shown by the broken line in Fig. 4. Because the average electron number decreases with increasing V concentration and the magnetic moments of V atoms are aligned antiparallel to the bulk magnetization, E_+ changes more rapidly than E_-, and Δ is reduced with increasing V concentration. These properties give rise to the increase of the exchange stiffness constant D as V atoms are added to Fe.

Cooke *et al.*[19] have been quite successful in calculating the spin-wave energies in various *d*-band metals including bcc Fe. Their calculations involve the *KKR* bands and two exchange parameters which are associated with the terms in the spin-dependent part of the Hamiltonian corresponding to the e_g and t_{2g} symmetries. These parameters are adjusted to reproduce the experimentally determined magnitude and symmetry character of the moment. In this scheme the exchange splitting is wave vector and band dependent in contrast to the simpler approach mentioned above. Using this more rigorous approach, the change in the spin-wave energy due to the addition of V atoms was estimated within the rigid-band approximation. The exchange parameters were assumed to be the same as those determined for Fe. The Fermi energy was adjusted so that the calculated moment agrees with the observed value for the average moment. For the alloy with the average magnetic moment of $1.95\mu_B$, which corresponds to $x\sim0.1$,[20] the calculated energies are shown in Fig. 5. Although the energies for small values of the wave vector do increase as observed, those for larger wave vectors are lower than those for Fe. This may be interpreted as an indication of the inadequacy of the rigid-band model.

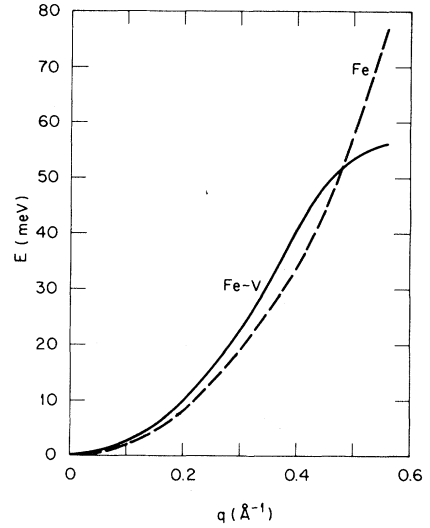

FIG. 5. Change in the spin-wave dispersion curve due to the addition of V atoms as calculated within the rigid-band model using the *KKR* bands and the exchange parameters for Fe. The solid line represents the calculated energy for the alloy with the average moment of $1.95\mu_B$ which corresponds approximately to $Fe_{0.9}V_{0.1}$.

V. CONCLUSION

A large amount of experimental data on the static magnetic properties of *d*-band-metal alloys have been accumulated in the past few years and they have been utilized to construct the band theoretic picture of the environmental effects on the magnetic moments in alloys. Although the data on the dynamical properties presented in this report can be interpreted within the Heisenberg model, the more fundamental interpretation based on the band theory seems more desirable, but that requires extremely time-consuming computer calculations associated with rigorous theoretical formulations. It is, therefore, quite important to acquire much more experimental data before serious efforts should be made in the theoretical calculations. We are currently studying the spin waves in the Ni-Pt alloy system.

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