PHYSICAL REVIEW B VOLUME 35, NUMBER 1 1 JANUARY 1987

Paramagnetic spin fluctuations in an $Fe_{65}Ni_{35}$ Invar alloy

K. Tajima
Department of Physics, Faculty of Science and Technology, Keio University, Hiyoshi, Yokohama 223, Japan

P. Böni and G. Shirane
Physics Department, Brookhaven National Laboratory, Upton, New York 11973

Y. Ishikawa* and M. Kohgi
Physics Department, Faculty of Science, Tohoku University Sendai 980, Japan
(Received 31 July 1986)

Polarized neutron scattering experiments have been performed on an $Fe_{65}Ni_{35}$ Invar alloy in the paramagnetic phase at $T=1.25T_c$. Constant-q spectra were fitted to simple Lorentzian functions and the q-dependence of the half-width Γ was found to agree with that of the spin-diffusion model in the measured q range ($0.1\ \text{A}^{-1}<q<0.9\text{A}^{-1}$). The amplitude of the spin fluctuation can be well described by the localized model with parameters which are deduced from the measured spin-wave dispersion at low temperature, the magnetization, and the uniform susceptibility. The model can be also applied satisfactorily to other itinerant metals such as Fe and Ni.

INTRODUCTION

The paramagnetic spin fluctuations in itinerant electron systems are still far from being well understood and have therefore been investigated extensively by neutron scattering techniques. As a typical example of an itinerant spin system, MnSi was studied in detail by Ishikawa *et al.*[1] The results are in agreement with the self-consistent renormalization theory proposed by Moriya and coworkers.[2] On the other hand, in localized spin systems, detailed experiments[3] on Pd_2MnSn were performed and the paramagnetic behavior over a wide temperature range could be understood rather well on the basis of the Heisenberg model with long-range interactions. Gd (Ref. 4) EuO (Refs. 5 and 6) and EuS (Ref. 7) were also studied as typical candidates for localized spin systems.

Fe and Ni, which are metals of particular interest in the field of magnetism, are intermediate cases for the moment localization and their magnetic properties are yet to be fully understood. Although paramagnetic scattering has been thoroughly studied, it is quite difficult to get sufficient information about the spin fluctuations in Fe (Refs. 8—10) and Ni (Refs. 11—14) because the scattering intensities extend to high-energy transfers, making it very difficult to obtain accurate intensity data. Recently, Brown *et al.*[15] and Shirane *et al.*[10] observed the paramagnetic scattering of Fe out to the Brillouin-zone boundary where it was found that the scattering cross section is confined to energies below approximately 60 meV.

$Fe_{65}Ni_{35}$ Invar alloy is considered to also be an intermediate case[16] between the localized and itinerant limits. FeNi alloy around this composition exhibits a substantial decrease of the thermal-expansion coefficient below the Curie temperature.[17] This is called the Invar effect and is the result of the compensation of the normal thermal expansion with the large magnetovolume effect associated with the variation of the amplitude of the local magnetic moment. The spin-wave energies[18] and the Curie temperature also decrease around this composition. Therefore, the neutron scattering intensities from the paramagnetic spin fluctuations are expected to be found in the low-energy region, which makes the measurement less difficult. In order to obtain detailed information for $Fe_{65}Ni_{35}$ in the paramagnetic region, a neutron scattering experiment has been performed above the Curie temperature using the polarized-beam technique.[19] The pioneering work of the paramagnetic scattering from the FeNi Invar alloy was made by Collins[20] more than 20 years ago using the time-of-flight method. The results were analyzed on the basis of the model of an ideal paramagnet. This model is too simple to give a correct description of the paramagnetism unless the temperature is far above the Curie temperature.

The present measurements were initiated partly because we expected to observe some anomalous properties of the paramagnetic scattering due to the Invar effect. We ended, however, with results which can be interpreted in terms of $\chi(q)$ based on the localized model.

EXPERIMENTAL RESULTS

The neutron scattering experiments were performed on a polarized-beam triple-axis spectrometer at the High Flux Beam Reactor at Brookhaven National Laboratory. $Cu_2MnAl(111)$ crystals were used both as a monochromator and analyzer. The constant-q data were mainly taken with a fixed final energy of $E_f=30.5$ meV although some of the data were taken with $E_f=40$ and 60 meV. The horizontal collimation was 40′-80′-80′-80′. The measurements were made along the [111] and [100] directions mainly around the (111) reciprocal lattice point. The paramagnetic scattering intensities were deduced by taking the intensity difference between the measurements mode with a horizontal field (HF) and a vertical field

(VF) present at the sample site with a spin flipper on. The single-crystal sample (~6 cm^3 in volume) was the same as that used for the spin-wave measurements.[18] The sample was mounted in a furnace and heated to 684 K, which is 136 K higher than the Curie temperature. Below this temperature, the beam was depolarized, probably due to surface inhomogeneities. Therefore, the half-width Γ was not observable at $T=T_c$.

The scattering function $S(q,\omega)$ above the Curie temperature can be expressed[11] in general as

$$S(q,\omega)=2k_BT\chi(q)\frac{\hbar\omega/k_BT}{1-\exp(-\hbar\omega/k_BT)}\frac{1}{\pi}\frac{\Gamma}{\Gamma^2+\omega^2}\ , \qquad (1)$$

where χ is the susceptibility. The amplitude of the spin fluctuation $\langle M(q)^2\rangle$ is given by the integration of Eq. (1) over energy,

$$\langle M(q)^2\rangle=6\int_{-\infty}^{\infty}S(q,\omega)d\omega\ . \qquad (2)$$

Examples of typical constant-q scans along the [111] direction are shown in Fig. 1. The solid lines in the figure are the result of fitting the data with the expression given by Eq. (1) after being convoluted with the resolution function. The experimental results are reproduced satisfactorily. Figure 2 presents the half-width Γ from Eq. (1) obtained by this procedure. The q dependence of Γ is isotropic with respect to the wave vectors in the [111] and [100] directions and can be written as $\Gamma=\Lambda q^2$ with $\Lambda=41$ meV Å^2 as illustrated by the dashed line in Fig. 2. Dynamical scaling theory predicts[21] the form $\Gamma=Aq^{5/2}f(\kappa_1/q)$ for the width. The dotted line in the figure represents this function with $A=146$ meV Å$^{2/5}$. This prediction clearly deviates from the experimental results for $q>0.3$ Å^{-1}.

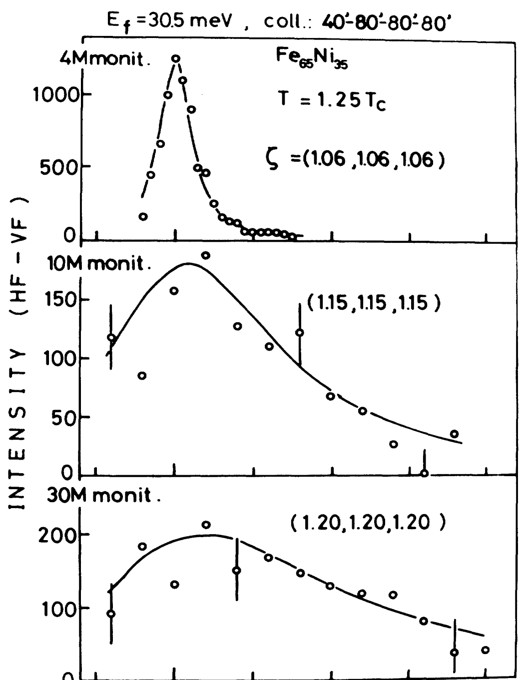

FIG. 1. Constant-q scattering data at $T=1.25T_c$ obtained using the horizontal field (HF)—vertical field (VF) technique. Solid lines are the results of fitting of Eq. (1) convoluted with the resolution function. 10M corresponds approximately 10 min counting time at $\hbar\omega=0$. Reduced wave vector ζ is given in units of $2\pi/d=3.04$ Å^{-1}, where d is the plane distance for the [111] direction.

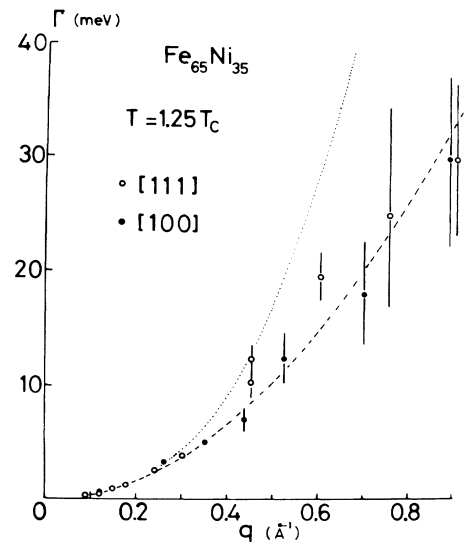

FIG. 2. The half-width Γ plotted against q for the [111] and [100] directions. The dashed line is a fit with $\Gamma=\Lambda q^2$ ($\Lambda=41$ meV Å^2). The dotted line represents $\Gamma=Af(\kappa_1/q)q^{5/2}$ with $A=146$ meV $q^{2/5}$.

$\langle M(q)^2\rangle$ in units of μ_B^2 was obtained from the measured constant q spectrum using the relationship[9]

$$\langle M(q)^2\rangle=\frac{I(q)}{0.0485f(Q)^2e^{-2W}}\ , \qquad (3)$$

where $I(q)$ is the integrated intensity of the constant-q measurement with respect to the energy transfer and is given in units of barns. $f(Q)$ is the magnetic form factor. The absolute value of $I(q)$ was determined by comparing the magnetic intensities with the integrated scattered intensities of several phonons the absolute values of which could be calculated.[9] $f(Q)$ of the FeNi Invar alloy determined by Ito *et al.*[22] was used in the present analysis. $\langle M(q)^2\rangle$ for the [111] and [100] directions thus obtained are shown in Fig. 3. They are isotropic in the measured q range. The width of the half maximum of $\langle M(q)^2\rangle$ in this figure gives the inverse correlation length $\kappa_1=0.24$ Å^{-1}. $\langle M(0)^2\rangle$ can be expressed as[23]

$$\langle M(0)^2\rangle=3k_BT\chi(0)\ , \qquad (4)$$

where $\chi(0)$ is the uniform susceptibility. Small pieces were sliced from the top and the bottom part of the crystal used for the neutron measurements and the susceptibility was measured at 684 K using a Faraday balance. Combining Eq. (4) with the value of $\chi(0)$ allows one to determine the value of $\langle M(0)^2\rangle$ shown in Fig. 3. The er-

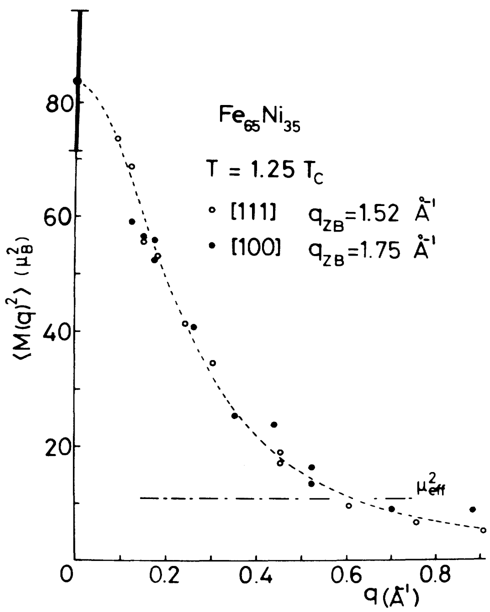

FIG. 3. The amplitude of the spin fluctuation $\langle M(q)^2\rangle$ defined by Eq. (2). q_{ZB} is the value of q at the Brillouin zone boundary. $\langle M(0)^2\rangle$ is obtained from the static susceptibility. The dashed line is the calculated curve using Eq. (8) (see the text). The dot-dash line represents ideal paramagnetic scattering with $\mu_{eff}=3.3\mu_B$.

ror bar corresponds to the difference in the susceptibilities obtained for the two slices. This difference may be caused by the inhomogeneity of the alloy concentration. The horizontal dot-dash line represents ideal paramagnetic scattering with an effective magnetic moment $\mu_{eff}=3.3\mu_B$ (Ref. 17).

DISCUSSION

One of the purposes of this study was to detect signatures which are directly related to the Invar effect. Unfortunately, we were unable to find any properties, either in the dynamics or statics, which can unambiguously be related to the Invar properties of $Fe_{65}Ni_{35}$. We believe, however, that this study provides important information concerning the paramagnetic scattering in itinerant spin systems.

$\chi(q)$ or $\langle M(q)^2\rangle$ can be calculated within the framework of the localized model. Using the spherical approximation[24] or Green's function method,[25] one can express $\chi(q)$ for the ferromagnet in the paramagnetic region as[26]

$$\chi(q)=\frac{(g\mu_B)^2}{\frac{(g\mu_B)^2}{\chi(0)}+2[J(0)-J(q)]} \quad (5)$$

$$=\frac{\chi(0)}{1+\frac{\hbar\omega_q\chi(0)}{(g\mu_B)^2S}}, \quad (6)$$

where $\hbar\omega_q$ is the spin-wave energy at low temperatures given as

$$\hbar\omega_q=2S[J(0)-J(q)]\ . \quad (7)$$

Therefore, $\langle M(q)^2\rangle$ can be expressed as[23]

$$\langle M(q)^2\rangle\cong 3k_BT\chi(q)=\frac{\langle M(0)^2\rangle}{1+\frac{\hbar\omega_q\langle M(0)^2\rangle}{3k_BT(g\mu_B)^2S}}\ . \quad (8)$$

Alternatively, Eqs. (5) or (8) can be derived by using a molecular field approximation[27] upon assuming a Curie-Weiss behavior for $\chi(0)$. This expression has been used previously in the analysis of localized spin systems[3] such as Pd_2MnSn. For a nonlocalized system such as the FeNi Invar alloy, Eq. (8) is also applied by using the measured spin-wave energies, $\langle M(0)^2\rangle$ and the saturation magnetization. The spin-wave dispersion relation has been obtained by Kohgi *et al.*[18] and is expressed as $\hbar\omega_q=Dq^2(1-\beta q^2)$ with $D=143$ meV Å^2 and $\beta=0.12$. The calculated $\langle M(q)^2\rangle$ is illustrated in Fig. 3 by the dashed line. Although there is no adjustable parameter in this calculation, the agreement with experiment is excellent.

The same method of comparison of $\langle M(q)^2\rangle$ can be also applied to Fe and Ni. The experimental results[10,12] and the calculations are illustrated in Fig. 4, together with the result[3] for Pd_2MnSn. Solid lines are the calculated curves obtained by using Eq. (8), which are also in good agreement with experiments. The parameters used in the

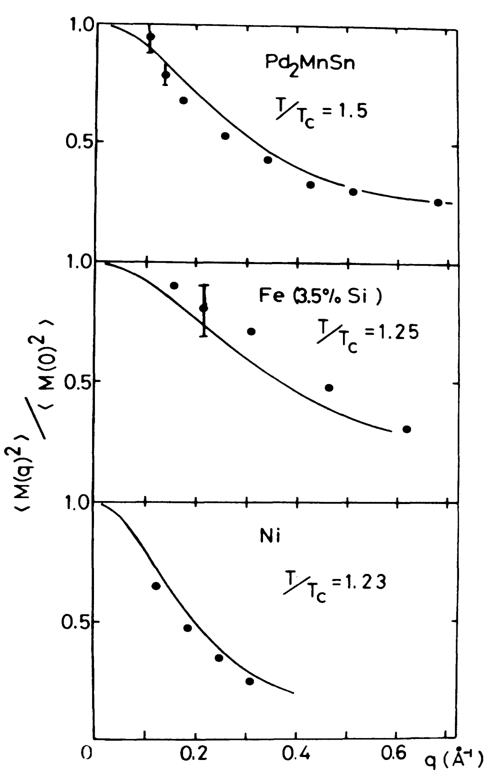

FIG. 4. $\langle M(q)^2\rangle/\langle M(0)^2\rangle$ of Pd_2MnSn, Fe, and Ni. Solid lines are the calculated results with Eq. (8) (see the text). In Pd_2MnSn, the observation at the largest q value corresponds to that of 80% to the zone boundary.

TABLE I. Parameters for the calculation in Eq. (8) obtained from measurements of the spin-wave dispersion, susceptibility, and saturation magnetization. Spin-wave measurements of Refs. 32 and 33 were made at room temperature while the measurements of the FeNi Invar alloy (Ref. 18) were made at 4.2 K. D and β of Fe (3.5% Si) are the estimated values from Ref. 32.

	$\hbar\omega_q = Dq^2(1-\beta q^2)$ D (meV) Å^2	β	$\langle M(0)^2\rangle$ μ_B^2	S
$Fe_{65}Ni_{35}$	143	0.12 (Ref. 18)	84	0.91
Fe (3.5% Si)	270	0.7 (Ref. 32)	55 (Ref. 10)	1.11
Ni	400	0 (Ref. 33)	15.5 (Ref. 12)	0.3

calculations are listed in Table I. Thus, Eq. (8), which is based on the localized model, describes very well the behavior of $\langle M(q)^2\rangle$, not only in localized spin systems, but in itinerant spin systems. Therefore, we conjecture that the spin fluctuations above T_c in both localized and itinerant spin systems are predominantly controlled by the exchange interactions, the magnetic moment, and the uniform susceptibility. It should be noted, however, that in a localized spin system such as Pd_2MnSn, Eq. (8) can be calculated for q out to the zone boundary and the result compares favorably with experiments as shown in Fig. 4. On the other hand, in the Fe, Ni, and FeNi Invar alloy, such a calculation can be applied only for q's less than half-way to the zone boundary, since a well-defined spin wave can be observed only in this q range. Recently, Mook and Paul[28] measured the spin-wave dispersion in Ni over much of the Brillouin zone. The spin wave in the large q region is of a greatly reduced intensity and very damped. Hence, the concept of a "spin wave" at large q in the itinerant spin system may be rather different from that in the localized spin system. Therefore, we speculate that the characteristic properties of the paramagnetic spin fluctuations in the itinerant spin system may appear at large q.

The q dependence of Γ was found to agree with that of the spin diffusion model $\Gamma = \Lambda q^2$ in a rather wide q range, as has been shown in Fig. 2. Initially, we considered this hypothetical formula as a convenient method of parametrization. Recently, a detailed account of the dynamic properties of the itinerant spin system in the paramagnetic region has been presented by Takahashi.[29] However, the spin diffusion behavior of Γ over a wide q region cannot be reproduced by this theory. In order to explain this behavior, it might be useful to take into account the influence of the impurity scattering of the electrons in the case of alloys such as FeNi. This was studied by Fulde and Luther[30] who showed that the spin diffusion behavior holds over a wide q range if the mean free path of the electrons is very small. The theory was extended by Fukuyama[31] with the use of a coherent potential approximation. Further experiments on dynamic behavior for other alloy systems are expected to shed more light on this important question.

ACKNOWLEDGMENTS

This work was carried out as part of the U.S.-Japan cooperative neutron scattering program. The authors would like to thank T. Moriya and Y. Takahashi for theoretical discussions, S. Anzai and S. Yuri for susceptibility measurements, and N. Wakabayashi and J. Z. Larese for a critical reading of the manuscript. Two of the authors (K.T. and Y.I.) appreciated the kind hospitality of the members of the neutron scattering group at Brookhaven National Laboratory. Work at Brookhaven National Laboratory was supported by the Division of Materials Science, U.S. Department of Energy (DOE) under Contract No. DE-AC02-76CH00016.

*Deceased.

[1]Y. Ishikawa, Y. Noda, Y. Uemura, C. F. Majkrzak, and G. Shirane, Phys. Rev. B **31**, 5884 (1985).
[2]See for example, T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer, Berlin, 1985).
[3]M. Kohgi, Y. Endoh, Y. Ishikawa, H. Yoshizawa, and G. Shirane, Phys. Rev. B **34**, 1762 (1986).
[4]J. Cable, R. Nicklow, and N. Wakabayashi, Phys. Rev. B **32**, 1710 (1985).
[5]P. Böni and G. Shirane, Phys. Rev. B **33**, 3012 (1986).
[6]H. Mook, Phys. Rev. Lett. **46**, 508 (1981).
[7]H. G. Bohn, K. Kollmar, and W. Zinn, Phys. Rev. B **30**, 6504 (1984).
[8]P. Brown, H. Capellmann, J. Deportes, D. Givord, and K. Ziebeck, J. Magn. Magn. Mater. **30**, 243 (1982).
[9]J. Wicksted, P. Böni, and G. Shirane, Phys. Rev. B **30**, 3655 (1984).
[10]G. Shirane, P. Böni, and J. Wicksted, Phys. Rev. B **33**, 1881 (1986).
[11]Y. Uemura, G. Shirane, O. Steinsvoll, and J. Wicksted, Phys. Rev. Lett. **51**, 2322 (1983).
[12]O. Steinsvoll, C. Majkrzak, G. Shirane, and J. Wicksted, Phys. Rev. B **30**, 2377 (1984).
[13]J. Martinez, P. Böni, and G. Shirane, Phys. Rev. B **32**, 7037 (1985).
[14]J. Lynn and H. Mook, J. Magn. Magn. Mater. **54-57**, 1169 (1986).
[15]P. Brown, H. Capellman, J. Deportes, D. Givord, S. Johnson, J. Lynn, and K. Ziebeck, J. Phys. (Paris) **46**, 827 (1985).
[16]Y. Ishikawa, J. Magn. Magn. Mater. **14**, 2025 (1979).
[17]M. Matsui, K. Adachi, and S. Chikazumi, J. Appl. Phys. **51**, 6319 (1980).

[18]M. Kohgi, Y. Ishikawa, and N. Wakabayashi, Solid State Commun. **18**, 509 (1976).
[19]K. Ziebeck and P. Brown, J. Phys. F **10**, 2015 (1980).
[20]M. Collins, Proc. Phys. Soc. (London) **86**, 973 (1965).
[21]R. Résibois and C. Piette, Phys. Rev. Lett. **24**, 514 (1970).
[22]Y. Ito, J. Akimitsu, M. Matsui, and S. Chikazumi, J. Magn. Magn. Mater. **10**, 194 (1979).
[23]The definition of $\langle M(q)^2 \rangle$ in this paper differs by a factor 4 from that in, for example, Ref. 10. This is because $\chi(q)$ in Ref. 10 is defined for $\mu_B^2 S(S+1)$, not for the magnetic moment $(g\mu_B)^2 S(S+1)$.
[24]S. Lovesey and R. Meserve, J. Phys. C **6**, 79 (1971).
[25]For example, K. Kawasaki, and H. Mori, Prog. Theor. Phys. **28**, 690 (1962).
[26]The equivalent equation can also be derived from the local moment limit of the band theory as is shown in Ref. 2.
[27]W. Marshall and R. Lowde, Rep. Prog. Phys. **31**, 705 (1968).
[28]H. Mook and D. Paul, Phys. Rev. Lett. **54**, 227 (1985).
[29]Y. Takahashi, J. Phys. Soc. Jpn. **55**, 2390 (1986).
[30]P. Fulde and A. Luther, Phys. Rev. **170**, 570 (1968).
[31]H. Fukuyama, Phys. Rev. B **5**, 2872 (1972).
[32]M. Collins, V. Minkiewicz, R. Nathans, L. Passell, and G. Shirane, Phys. Rev. **179**, 417 (1969); H. Mook and R. Nicklow, Phys. Rev. B **7**, 336 (1973).
[33]V. Minkiewicz, M. Collins, R. Nathans, and G. Shirane, Phys. Rev. **182**, 624 (1969).