"Comment on 'Spin-wave resonance studies in Invar films' "-A reply

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In this reply the author challenges the suggestion by Wohlfarth that the samples used in this study were anomalous. It is emphasized that annealed film samples have magnetic properties consistent with bulk materials. The composition dependence of the dispersion coefficient, observed to have a minimum at about 40-at. % Ni, is consistent with observations by other laboratories reporting spin-wave resonance measurements and small-angle neutron scattering measurements.

In the Comment by Wohlfarth¹ (EPW) the suggestion is made that the films used in our study² (BW) are anomalous and that the value of our results is questionable. We argue against that conclusion as follows.

The study of standing spin-wave resonance in thin magnetic films has revealed that a number of difficulties that can introduce errors into the data analysis may arise in film preparation. These difficulties included inhomogeneity in the internal field, the precise nature of the boundary conditions at the film surfaces, and the sources contributing to the internal field. In our paper, it was the intent to remove as many of these ambiguities as possible. This was done by determining the dispersion relation from the higher-order spin-wave modes where the influence of the inhomogeneities is minimal and by quoting values of the internal field rather than the demagnetization field $4\pi M$. The magnetic resonance is influenced by anisotropy fields and stress-induced magnetostriction fields as well as by the demagnetization field, and it is not possible to distinguish between them. If the internal field has the value expected from the demagnetization field, the two are often assumed to be equivalent.

In Fig. 12 of the original report for our work (BW), data are recorded for three kinds of films: films having only bcc crystal structure, films having only fcc crystal structure, and a third type of film having regions of bcc and fcc structure. The internal fields reported for the mixed samples correspond to that phase having the larger magnetization, i.e., the bcc phase. Equating the observed internal field to $4\pi M$ for the bcc phase, a continuous curve (solid black dots and open squares) is observed at room temperature. These values are in reasonable agreement with bulk sample magnetization in the region below the Invar point. The magnetization in this region is not expected to reduce to zero at the Invar point, since that question is related to the fcc crystal structure, not the bcc crystal structure.

The third set of data reported in Fig. 12 of BW

is related to films that have fcc structure only. This is evidenced by the single set of resonance modes and by the presence of only the fcc x-ray diffraction line for these films. For those films having more than 40-at. % nickel in iron, this internal field is again in reasonable agreement with $4\pi M$ of bulk materials. For fcc films having less than 40-at. % nickel, the internal fields measured in the films as deposited, i.e., with no annealing, is considerably larger than the values of $4\pi M$ determined from measurements in bulk samples.³ These values are represented by the diamond indicators in Fig. 12 of BW. However, when these fcc films are annealed, the internal field is observed to decrease considerably to where the internal-field values are *consistent* with the values of $4\pi M$ reported for bulk samples. Such bulk measurements have been made by Bando,³ who reported that at 40-at. % Ni the bulk $4\pi M$ values are slightly larger than 15 kG, that at 35-at. % Ni this value has reduced to about 12.5 kG, and that at 30-at.% Ni the magnetization is near zero. In Fig. 12 of BW, points were not reported for samples having less than 33-at. % Ni because such films had a mixed crystal structure, and the effect of the regions having bcc structure on the internal fields of the fcc material was not certain. During the annealing process, the internal field of the films was also observed to become more homogeneous as evidenced by the decrease in the intensity of the higher-order spin-wave modes. However, there is no evidence that D is rapidly decreasing as the Invar point is approached.

Over the range of concentrations considered, our results are in good agreement with those in Ref. 4, where a detailed comparison between experiments is made. Thus it still remains the conclusion of the authors that the fcc films being investigated do have the sharp drop in the magnetization characteristic of bulk samples and that our samples do not represent anomalous specimens.

Before turning to the discussion of the spin-wave dispersion coefficient, the authors wish to point out two errors in the original manuscript.² One is

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that the value of the exponent in the upper-lefthand side of Figs. 10 and 11 should read 10^{-29} instead of 10^{-27} . Second, the figure captions are reversed for Figs. 10 and 11.

For those points having more than 35-at. % nickel, the data plotted in Fig. 10 of BW correspond to the values obtained from the annealed films having fcc phases only. These samples indicate a definite minimum in the observed D value near 45-at. %nickel, where the $4\pi M$ values of the films are in excellent agreement with the values reported for the bulk material. It is interesting to note that the minimum in D occurs at the maximum value of Mobserved for the FeNi system. The two sets of Dvalues reported for samples having less than 35at. % nickel were obtained from samples deposited over a thin gold film. These values are considerably larger than those for higher concentration of nickel, and it may be due to an uncertainty in the boundary condition at the alloy-gold interface. However, these points are consistent with the trend that as the Invar point is approached, an increasing value of D is observed for the fcc phases having mixed crystal structures.

EPW also refers to other measurements of D. These data are reproduced in Fig. 1. While there is a considerable scatter in the data observed in the various laboratories, any one set of data can be represented by a reasonably smooth curve which shows a minimum in D at concentrations between 40- and 50-at. % nickel. This suggests that D is increasing as the Invar region is approached. The only exception to this minimum is the small-angular-scattering data of Hatherly *et al.*⁵ These data were obtained from samples having a minimum concentration at 40-at. % nickel, and at that concentration the value of D reported is consistent with the values observed in the spin-wave resonance spectra of the films.

The solid line in Fig. 1 is a theoretical curve obtained from Katsuki's plot of D vs T_c obtained for the rigid-band model.⁶ A reasonable agreement is observed for nickel concentrations larger than 40-at.%. The experimental results reported by the three different laboratories that have obtained data for concentrations below 40-at.% nickel indicate that the D value deviates dramatically from the predictions of the model.

An additional dimension to this question has been raised by recent measurements of the temperature dependence of the magnetic excitations in nickel.⁷ These experiments, performed at temperatures ranging from 4.2 to 715 K, indicated that the spin waves change very little in character even at 80° above the Curie temperature. At about 80 meV, a sudden decrease in the spin-wave intensity was interpreted as the intersection of the spin-wave spectrum with the continuum of the Stoner excitations.

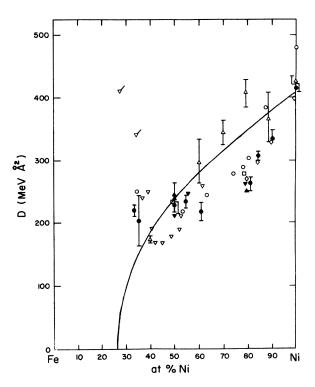


FIG. 1. Plot of the *D* value vs the composition of Ni in fcc Fe-Ni alloys as reported by several groups using the following experimental techniques: from spin-wave resonance: ∇ , Bauer and Wigen, Ref. 2; \bigoplus , Maeda, Yamauchi, and Watanabe, Ref. 4; O, Rusov, Ref. 8; \checkmark , Weber and Tannenwald, Ref. 9; from neutron diffraction: Δ , Hatherly *et al.*, Ref. 5; \blacktriangle , Menzinger *et al.*, Ref. 10; from the measurements of the magnetization at low temperatures: \Box , Argyle and Charap, Ref. 11; \Diamond , Maeda, Yamauchi, and Watanabe, Ref. 4. The solid line is a theoretical curve obtained from Katsuki's plot of *D* vs *T_c* obtained from the rigid-band model (Ref. 6).

The position of this intersection is also found to be quite temperature independent, suggesting that the Stoner gap is not decreasing to zero as the magnetization goes to zero. These results tend to suggest that the exchange splitting may not be related to the magnetization or the Curie temperature in ferromagnetic metals as is usually assumed in the itinerant-electron models.⁶

The authors are aware of many difficulties in the analysis of the thin-metal-film spin-wave resonance spectra. The effects of inhomogeneities and boundary conditions can introduce errors of several percent in the value of the demagnetization field and in the spin-wave dispersion coefficient but the general trend of all of the data from many laboratories is consistent with a nonzero dispersion coefficient at the Invar point.

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- ¹E. P. Wohlfarth, preceding paper, Phys. Rev. B <u>10</u>, xxxx (1974).
- ²C. A. Bauer and P. E. Wigen, Phys. Rev. B <u>5</u>, 4516 (1972).
- ³Y. Bando, J. Phys. Soc. Jap. <u>19</u>, 237 (1964).
- ⁴T. Maeda, H. Yamauchi, and H. Watanabe, J. Phys. Soc. Jap. <u>35</u>, 1635 (1973).
- ⁵M. Hatherly, K. Hirakawa, R. D. Lowde, J. F. Mallet, M. S. Stringfellow, and B. H. Torrie, Proc. Phys. Soc. Lond. <u>84</u>, 55 (1964).
- ⁶A. Katsuki and E. P. Wohlfarth, Proc. R. Soc. Lond. <u>295</u>, 182 (1966); A. Katsuki, Br. J. Appl. Phys. <u>18</u>,

er is referred to his paper⁴ where a more detailed comparison of the data is reported.

, 199 (1967).

- ⁷H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. <u>30</u>, 556 (1973).
- ⁸G. I. Rusov, Fiz. Tverd. Tela <u>9</u>, 196 (1967) [Sov. Phys.-Solid State <u>9</u>, 146 (1967)].
- ⁹R. Weber and P. E. Tannenwald, Phys. Rev. <u>140</u>, A498 (1965).
- ¹⁰F. Menzinger, G. Caglioti, G. Shirane, and R. Nathans, J. Appl. Phys. 39, 455 (1968).
- ¹¹B. E. Argyle and S. H. Charap, J. Appl. Phys. <u>35</u>, 802 (1964).