Spin-Wave Dispersion in Ferromagnetic Ni and fcc Co†

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Inelastic scattering of polarized neutrons by the diffraction technique from Ni and fcc Co at room temperature have yielded constants in the dispersion relation of the spin-wave energy versus wave vector, $\hbar\omega = Dq^2(1-\beta q^2)$. The results for fcc Co ($D=384\pm20$ meV Å^2, $\beta=3.1\pm1.0$ Å^2) indicate that this metal is similar to Fe and hcp Co in showing a quartic term even for relatively small wave vector ($q/q_{\max}\lesssim0.15$). In Ni, on the other hand ($D=374\pm20$ meV Å^2, $\beta=0\pm1.0$ Å^2), a more nearly quadratic relation is found in the same range of q. It is argued that these results, when compared with others for Fe and Ni-Fe alloys, are inconsistent with an attempt to interpret the magnetism of these metals in terms of a generalized Heisenberg model with long-range interactions.

I. INTRODUCTION

THE spin-wave dispersion constant for ferromagnetic metals may be directly measured by two methods: thin-film resonance and inelastic neutron scattering. The first of these, restricted to long wavelengths, determines most accurately the leading quadratic term in the dispersion relation; the second, although inaccurate at small wave vector because of resolution problems, potentially can determine the complete dispersion curve. Investigations have been carried out on Ni and fcc Co by both these methods. Thin-film spin-wave resonance measurements were performed by Tannenwald and Weber,[1] Kimura and Nosé,[2] Frait and Ondris,[3] and Phillips and Rosenberg.[4] Neutron-scattering measurements with energy analysis were carried out by Sinclair and Brockhouse[5] on Co, while Hatherly *et al.*[6] have reported small-angle scattering measurements on Ni.

A special application of the neutron method, namely, inelastic scattering of polarized neutrons, is particularly well suited for investigation of ferromagnetic spin waves. This is because suitable correlation of the neutron polarization, magnetization, and scattering vectors unambiguously isolates the spin-wave scattering from other inelastic components.[7] Even without energy analysis, this method (the so-called diffraction method) has proved useful in the range of intermediate magnon wave vectors ($0.05\lesssim q/q_{\max}\lesssim0.3$) when (as is usually the case) there exists a single-valued dispersion curve that gives rise to closed scattering surfaces in reciprocal space.

Previously, the diffraction method was used[8,9] to demonstrate that the spin-wave dispersion curves in ferromagnetic Fe and hcp Co deviate from a quadratic form even for relatively small wave vectors. In this paper, extensions of earlier preliminary results[10] on fcc Co and Ni are presented which indicate that fcc Co is similar in behavior to the above-mentioned elements, but there is no appreciable quartic term in the dispersion law for Ni. Thus, of all the $3d$-transition metals investigated so far in this manner, Ni is alone in showing a very nearly quadratic relation between spin-wave energy and wave vector.

II. EXPERIMENTAL DETAILS

Measurements were made at incident-neutron wavelengths of 1.05–1.07 Å and at room temperature. In both cases, the crystals were magnetized along a ⟨100⟩ direction in the (011) plane by a field of approximately 3000 Oe, and spin-wave propagation was observed in this plane near the (200) reciprocal lattice point. Under these conditions, the propagation direction varies for small momentum transfer, but here the scattering is isotropic; for larger momentum transfer, where anisotropy of the dispersion might be expected, the propagation vector is nearly along [100].

The crystal of Co was an alloy containing 6 at.% Fe (to stabilize the fcc phase) in the shape of square slabs, 2 mm in thickness and 1–1¼ in. on a side. In the case of Ni, a single crystal of the isotope Ni^{60} was grown, in order to reduce the isotopic spin disorder scattering

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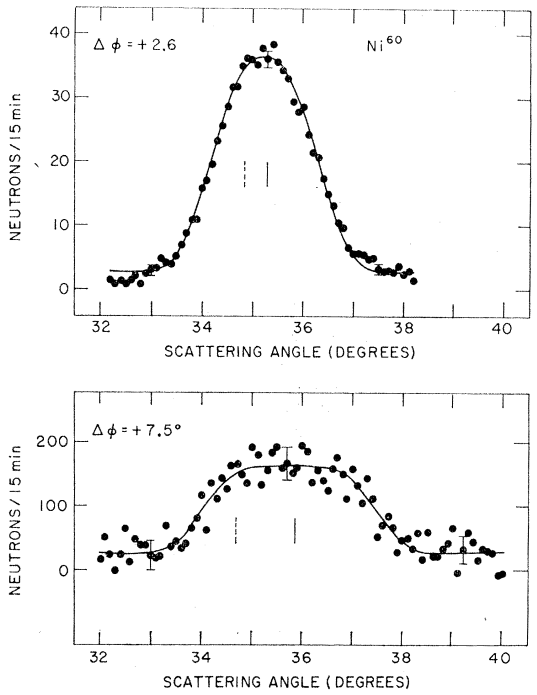

FIG. 1. Spin-wave scattering near the (200) reflection in Ni^{60} as a function of detector angle for two crystal misset angles ($\Delta\varphi$). The dashed bar represents the position of the Bragg peak, the solid bar the expected center of the distribution calculated from geometrical considerations.

background and, by virtue of the smaller coherent nuclear scattering amplitude, the scattered phonon intensity as well. These contributions were the main cause of the statistical uncertainty that plagued the earlier measurements. Two Ni^{60} crystals, cut from the same boule and oriented side by side, were used; they were of cylindrical shape, $\sim\frac{3}{8}$ in. in diameter, and $1\frac{1}{4}$ in. long. The bulk of the measurements were made at the Brookhaven graphite reactor with some final data being taken at the high flux beam reactor.

The experimental method, which has been fully described elsewhere,[11] consists in determining the width of the inelastic scattering surfaces as the crystal is misset at increasing angles away from the Bragg position, thus allowing progressively larger energy transfers. Since the scattered-neutron distribution is seen by a system of finite vertical and horizontal resolution, the true angular width is convoluted with an experimental resolution function. In the present case, the width was extracted by a computer program which convolutes the ideal square shape appropriate to a rotationally symmetric surface with a resolution function determined from the Bragg peak; the result is then least squares fitted to the observations, with the width of the surface as a parameter. The resolution function used is believed to be a good approximation as long as the position of the wings of the observed distribution does not depart much from the focusing position and its width is less than the vertical acceptance angle of the detector (about 6° in the present case).

In the present investigation, measurements were made for missets of up to 30° for Ni and 25° for Co, although missets for which the above condition on the width was not met were not used in the analysis. Missets were taken in the direction of both increasing and decreasing scattering angle corresponding to spin-wave creation and annihilation, respectively. Typical scans for Ni and fcc Co are reproduced in Figs. 1 and 2, where the difference count for neutron spin parallel and antiparallel to the field is plotted, representing the spin-wave cross section in isolation. The points are the raw data and the smooth curve is the least-squares-fitted function mentioned above.

III. ANALYSIS OF THE RESULTS

From the relations between momentum and energy conservation and the observation of a cutoff in the scattered magnon intensity, one has the relation[11]

$$k_f=\frac{-k_1\cos(\Gamma/2)\epsilon g(q^2)}{1-\epsilon g(q^2)},$$

with

$$g(q^2)=\frac{\hbar^2}{4mq}\frac{dE(q^2)}{dq} \quad \text{and} \quad \mathbf{k}_1=\mathbf{k}_i+2\pi\boldsymbol{\tau},$$

where $E(q^2)$ is the dispersion relation, $\mathbf{k}_i$ and $\mathbf{k}_f$ are the incident- and final-neutron wave vectors, $\boldsymbol{\tau}$ is a recipro-

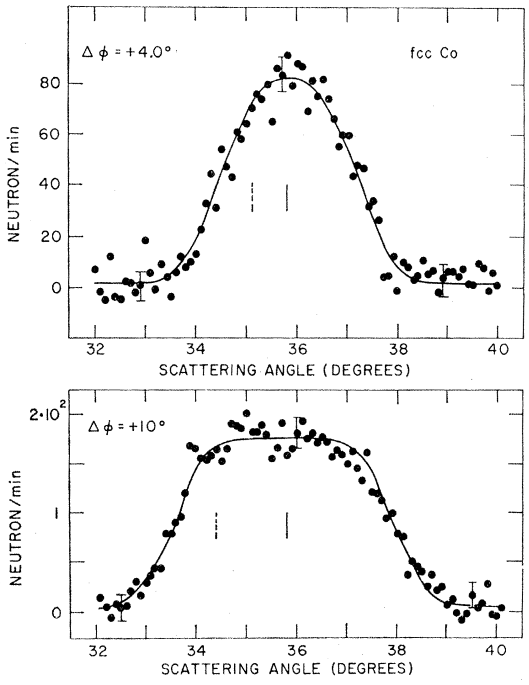

FIG. 2. Same as Fig. 1, for Co.

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TABLE I. Spin-wave dispersion constants for Ni and fcc Co.

		Ni	fcc Co
One-parameter fit	D (meV Å^2)	374	356
	$\bar{\sigma}$ (meV)[a]	1.9×10^{-4}	1.0×10^{-4}
Two-parameter fit	D (meV Å^2)	391	384
	β (Å^2)	1.02	3.13
	$\bar{\sigma}$ (meV)	1.9×10^{-4}	0.75×10^{-4}

[a] $\bar{\sigma}$, the average standard deviation of the experimental points from the fitted curve, is defined as $[\Sigma_n(E_{\text{calc}}-E_{\text{obs}})^2]^{1/2}/n$, where E_{obs} are the n observations transformed to (E,q) space.

cal lattice vector, Γ is the width of the scattered-neutron distribution, and $\epsilon=+1$ or -1 for neutron energy gain or loss, respectively.

For a quadratic dispersion relation, $E=E_0+Dq^2$, a linear relation exists between $\sin^2(\Gamma/2)$ and $(k_i/k_1)^2$ with the slope proportional to D^{-1} and the intercept to E_0. In the case of Fe, the observation of a nonlinear relationship between these quantities suggested a generalization of the above relation, still assuming rotational symmetry of the scattering surface about k_f. In this generalization, the dispersion curve is expanded in even powers of q, and the observed relation between $\sin^2(\Gamma/2)$ and $(k_i/k_1)^2$ is used in an iterative procedure to construct a dispersion curve that is least-squares fitted to the observations [transformed to (E,q) space] with the coefficients of the powers of q^2 as parameters. Using this method of analysis, a much better statistical fit was obtained[8,9] in the case of Fe and hcp Co when a fourth-order term was added to the dispersion relation.

If our present results for Ni and fcc Co are plotted in the form $\sin^2(\Gamma/2)$ versus $(k_i/k_1)^2$, they appear as in

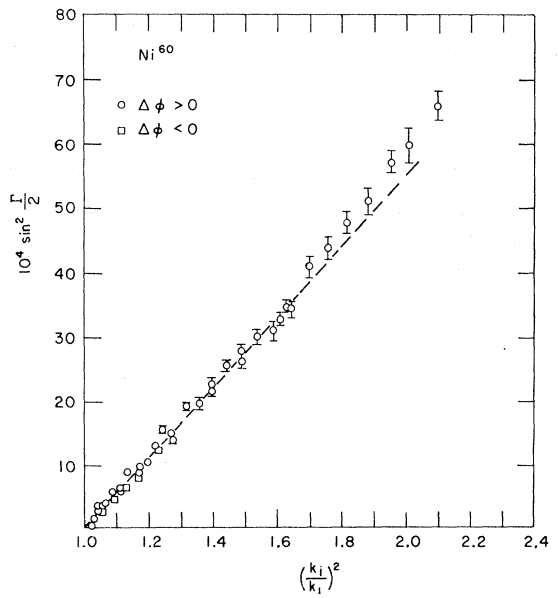

FIG. 3. Plot of $\sin(2\Gamma/2)$ versus $(k_i/k_1)^2$ for Ni^{60}. The dashed line corresponding to $D=374$ meV Å^2 was not fitted directly to these data, but by the method described in the text. $\Delta\varphi<0$ and $\Delta\varphi>0$ corresponds to neutron energy gain and loss, respectively.

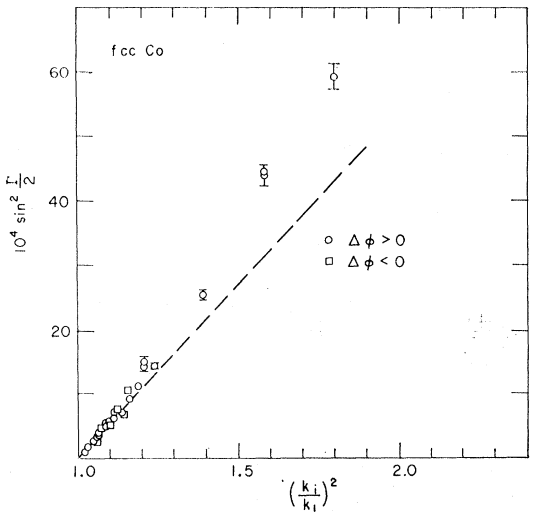

FIG. 4. Same as Fig. 3, for Co. The deviation of the data from the dashed line ($D=384$ meV Å^2) is a consequence of the quartic contribution, as discussed in the text.

Figs. 3 and 4. In both cases, the intercept is small, and we estimate that E_0 is $\lesssim0.5$ meV. If we now use the above-mentioned iterative procedure for reducing the data to a dispersion curve, we find the results shown in Table I when fitting with one paarmeter (quadratic term only) or with two parameters (quadratic plus quartic term). These fits were obtained for $q\leq0.26$ Å (missets $\leq17.5°$ in Ni and $\leq15°$ in Co) to an expression of the form $E=Dq^2(1-\beta q^2)$.

We note from the table that, for Co, significant improvement is obtained by including a quartic term, as was found to be true in the previous analysis[8,9] of Fe and hcp Co. In the case of Ni, either form, quadratic only or quadratic plus quartic, fits the data equally well, but the quartic term, surprisingly enough, is small relative to Co and Fe.

IV. DISCUSSION

We note at the outset that in none of our data have we been able to observe anomalies of the type reported[12,13] for these materials previously, which were inferred from discrepancies with a trapezoidally shaped expected neutron distribution. They were attributed to Kohn-type singularities in the s-d exchange contribution to the spin-wave energy, arising from the contact of the doubled Fermi surface with the spin-wave scattering surface. Examination of our data, such as is shown in Figs. 1 and 2, which was taken with better resolution than the earlier data, and in the case of Ni with considerably better signal-to-noise ratio, reveals, in no case, deviations larger than statistical error from

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TABLE II. Room-temperature dispersion constant D (in meV Å^2) for ferromagnetic metals.

Method \ Material	Fe	fcc Co	hcp Co	Ni
Neutron scattering	266±15[a] 256±8[b]	384±20[c] 371[f]	490±20[h]	374±20[e] 403±7[b]
Thin-film resonance	312±13[c] 281±17[d]	340[g]		354[g] 387±12[i]

[a] Reference 8.
[b] M. Stringfellow (private communication).
[c] T. Phillips, Phys. Letters **17**, 11 (1965).
[d] Z. Frait and M. Ondris, Phys. Status Solidi **2**, K185 (1962).
[e] Present work.
[f] Reference 5.
[g] Reference 4.
[h] Reference 9.
[i] Reference 2.

the least-squares-fitted expected shape. Therefore, we conclude that, if such anomalies exist, they are much smaller than have been claimed.

A systematic form-factor dependence in both fcc Co and Ni data was noticed, however, at the larger missets where the angular width was large. As in the case of Fe, this form factor suggests that the spatial distribution of the electrons participating in the spin-wave excitations is qualitatively $3d$-like in form.

The D parameters obtained by various methods on the ferromagnetic metals are intercompared in Table II. (Several thin-film results were omitted here because the magnetization was substantially less than that of the bulk material.) Where the same metal has been measured by more than one technique, reasonable agreement exists between the values when the probable errors of the measurements are taken into account. Such differences as do exist between the various methods do not appear to be systematic.

A β value for fcc Co may be derived from the dispersion curve measured by Sinclair and Brockhouse[5] by means of the three-crystal method. The cosine function used to fit their data, which extend to $q \simeq 0.36$ Å^{-1}, has $\beta = a_0^2/36 = 0.35$ Å^2. The origin of this difference could well lie in the data for small wave vector, where we believe the diffraction method is more accurate. In this region, the vertical component makes the effective wave vector in the three-crystal method larger, thus leading to a spurious energy gap and a smaller apparent D value, whereas in the diffraction method, the vertical components are measured and allowed for in the fitting process. However, it is important to point out that our values of D and β provide the best power series fit only for the measured range of q. Since the range for which such an expansion is valid is not known, it may be risky to extrapolate them for comparison with the three-crystal data taken at higher q.

Previously, the observation of a quartic term in Fe by neutron diffraction and in Ni_3Fe by thin-film resonance was interpreted,[8,14] on the basis of a generalized Heisenberg model, as an indication of a long-range exchange interaction in these materials. In particular, weight was placed[15] on the fact that by a 20% decrease in the value of D obtained from neutron scattering, the ratio of the effective range divided by the near-neighbor distance ($\langle\rho^2\rangle/r_1^2$) could be made to be of similar magnitude (≈ 120) in the two materials.

It is our belief that the present results for Ni are inconsistent with this interpretation. First of all, we obtain from our measurements on the basis of this procedure an effective-range parameter in Ni of $\langle\rho^2\rangle \approx 3r_1^2$, in contrast to the expectation that the effective range of Ni_3Fe should be nearer to that of Ni than of Fe. Moreover, if we assume that conduction electrons via the s-d interaction are the means of propagating the long-range interaction, there exists a body of evidence from conductivity, optical, and other measurements which suggests that the localized model is, if anything, less appropriate in Ni than in Fe.

Recent band-structure calculations[16] on Ni predict a dispersion curve for $q \lesssim 0.36$ Å^{-1} with a ratio of fourth- to second-order terms about twice the maximum value consistent with the present data. Whether or not this discrepancy is merely quantitative is difficult to say in the absence of equivalent calculations for Fe and Co. One would like to see such calculations predict relatively larger quartic contributions than those for Ni, to provide a clue to the observed difference in behavior.

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