EFFECT OF DILUTE SOLID SOLUTIONS OF IRON AND OF NICKEL ON THE NUCLEAR RESONANCE OF Co^{59}†

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In 1959 Gossard and Portis observed the first nuclear resonance signals from ferromagnetic Co^{59}, thus initiating a technique particularly suited to the study of the electronic structure of ferromagnets.[1]

In hope of obtaining additional information about the ferromagnetic state, we have studied the nuclear resonance spectra of the Co^{59} matrix in ferromagnetic solid solutions containing small amounts of iron or nickel. The experimental results are displayed in Figs. 1 and 2. In pure cobalt four lines are observed other than the principal resonance from nuclei in the fcc phase at 213.1 Mc/sec. The resonance at 221.3 Mc/sec is produced by nuclei in the hexagonal phase (both hexagonal and fcc phases of cobalt co-exist at room temperature).[2] The other three lines (218.5, 215.6, and 210 Mc/sec) arise through the interaction of nuclei in the fcc phase with stacking faults.[3] This interpretation is supported by the behavior of the spectra upon the addition to the cobalt matrix of iron, which stabilizes the fcc phase. First the line at 221.3 Mc/sec disappears as the concentration of iron is increased; then the other three disappear at slightly higher iron concentrations.

The addition of 1 or 2% iron to the cobalt matrix produces three new resonance lines, two higher in frequency than the pure cobalt resonance (215.6 Mc/sec and 214.2 Mc/sec) and one lower (210.1 Mc/sec).

The intensities of these lines are in the ratio

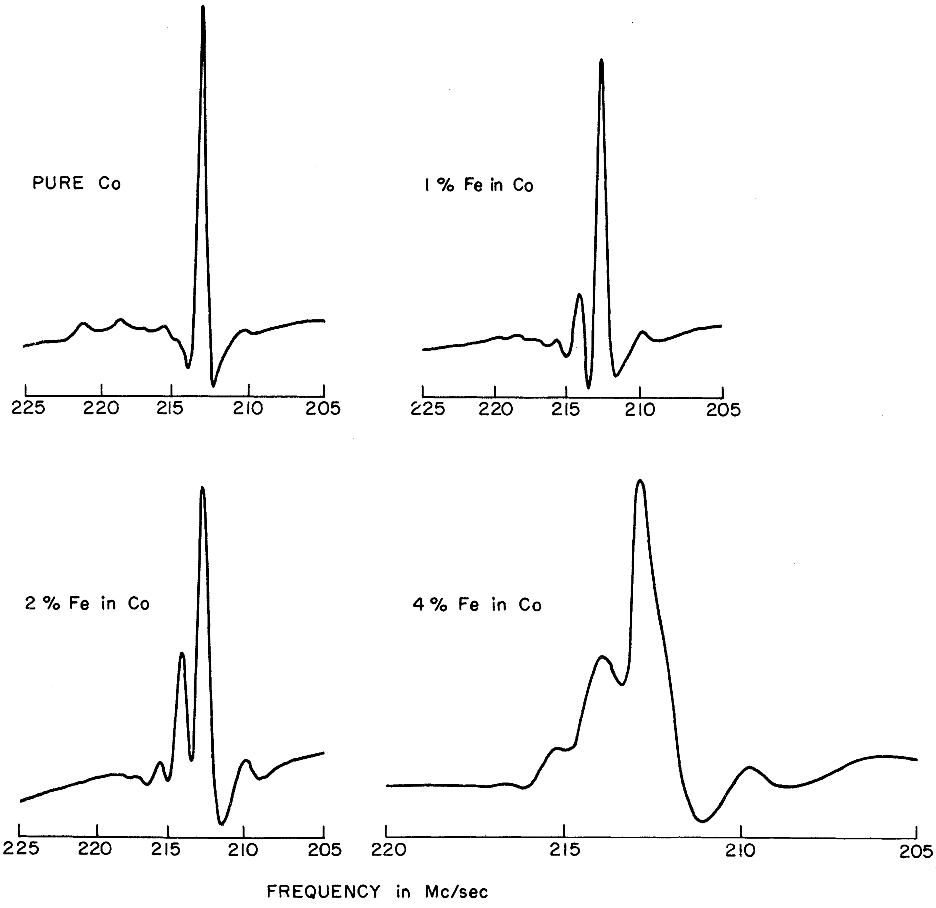

FIG. 1. The first derivative of the Co^{59} nuclear resonance spectra in pure Co and Fe-Co alloys as a function of frequency. The vertical scales are not comparable between spectra.

of 2:1:4 which strongly suggests that they arise from nuclei in shells about an iron impurity (there are twelve nuclei in the first shell, six in the second, and twenty-four in the third).

For the dilute nickel solutions the situation is much the same with respect to the effect of stacking faults and the resonance from the hexagonal phase. Stacking-fault lines are observed at higher concentrations of nickel than of iron, which is reasonable since nickel does not stabilize the fcc phase so well as iron.[4] Co^{59} resonance lines are found at 205, 209.5, 211.1, and 211.8 Mc/sec due to the presence of nickel. There is so much overlap of lines, however, that relative intensity measurements cannot be made as reliably as in the case of iron-cobalt alloys. Although intensity comparisons are difficult to make, we estimate that the total integrated intensity does not change upon alloying.

In trying to understand these results there are two general effects that one must consider. The first is the possibility of a direct interaction between an impurity atom and a nearest neighbor matrix atom. We can obtain some estimate of the size of this interaction from the low-temperature specific heat data.[5] For small concentrations of cobalt in iron the hyperfine field at the cobalt nucleus is increased by about 80 koe. Remembering that there are eight nearest neighbors in the bcc phase and assuming the interaction to be additive leads to an estimate of 10 koe for the effect of an iron atom on a cobalt nearest neighbor. This means that the nearest neighbor line is expected to be about 10 Mc/sec higher in frequency. It is observed 3 Mc/sec lower, however.

The second general effect is the scattering of conduction electrons by an impurity atom to produce a spin polarization about the impurity center. Ruderman and Kittel and later Yosida[6] showed that conduction electrons scattered by a localized spin are thereby spin polarized. The spin polarization is a periodic function of the radial distance from the scattering center and dies out as $1/(rk_f)^3$, where k_f is the value of the quasi-momentum at the Fermi surface. Friedel *et al.*,[7] considering the possibility of d electrons being in virtual bound states, have shown that in this case also the scattered conduction electrons will be spin polarized, the polarization again dying out as $1/(rk_f)^3$. We should like to point out that the scattering of conduction electrons from a localized charge[8] is sufficient to produce a spin polarization about the scattering

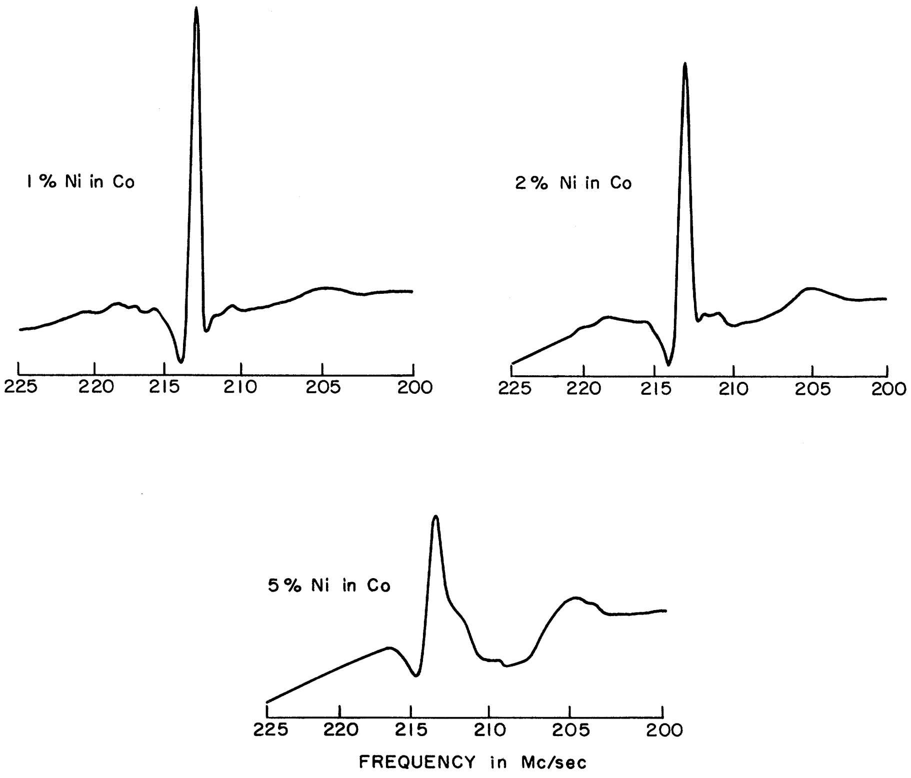

FIG. 2. The first derivative of the Co^{59} nuclear resonance spectra in Ni-Co alloys as a function of frequency. The vertical scales are not comparable between spectra.

center. Charge scattering causes a periodic variation in electron density about the localized charge, but in a ferromagnet this is equivalent to a variation in spin polarization since the conduction electrons are spin polarized through interaction with d electrons.[9] This polarization dies out as $1/(rk_f)^2$ and might therefore be expected to give the largest contribution to the effective magnetic field at distant neighbors.

We have attempted to fit the iron-cobalt alloy data by a combination of a direct interaction acting on the impurity nearest neighbors and charge scattering producing an effective field for all neighbors. We find that the data can be fitted rather well with a value of the quasi-momentum vector halfway to the edge of the first zone and a value of the s - d exchange energy corresponding roughly to the free-atom value.[9] Although the physical situation is not so simple as this, the results are quite encouraging. For the nickel alloys we find that charge scattering would predict the second and third neighbor lines shifted to lower frequency while the nearest neighbor line should be found at higher frequency. Although we cannot make a unique assignment of resonance lines to nuclear positions, we do observe all the lines shifted to lower frequency. This means that the nearest neighbor resonance position is not correctly predicted by scattering alone. But arguing again from the specific heat data,[5] we find that the direct interaction should shift the nearest neighbor line to lower frequencies.

The effect of stacking faults may also be treated as a scattering problem. A stacking fault in fcc Co is a region about 5000 atoms long, perhaps 200 atoms wide, and exactly 2 atoms thick, hexagonally packed. This region appears, so far as the conduction electrons are concerned, as a sheet of material of different propagation constants from the bulk fcc matrix. The interference of the incident and reflected electrons produces a periodic variation in electron density near the stacking fault that dies out inversely with the distance. The relative positions of the stacking fault lines agree qualitatively with the previous assignment of k_f, while the sizes of the frequency shifts are accounted for if the conduction electrons change effective mass by a few percent in crossing the stacking fault boundary.

A more detailed discussion of these various effects together with additional experimental results will be published elsewhere.

It has very recently come to our attention that work similar to ours is being done by Y. Koi and co-workers. We understand that their observations are generally similar to those reported here.[10]

We are pleased to express our thanks to Professor A. M. Portis for many interesting discussions.

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[1] A. C. Gossard and A. M. Portis, Suppl. J. Appl. Phys. 31, 205S (1960).

[2] Y. Koi, A. Tsujimura, and T. Kushida, J. Phys. Soc. Japan 15, 2100 (1960).

[3] R. Street, D. S. Rodbell, and W. L. Roth, Phys. Rev. 121, 84 (1961); W. Hardy, J. Appl. Phys. (to be published).

[4] M. Hansen, Constitution of Binary Alloys (McGraw-Hill Book Company, Inc., New York, 1958).

[5] V. Arp, D. Edmonds, and R. Petersen, Phys. Rev. Letters 3, 212 (1959).

[6] M. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954); K. Yosida, Phys. Rev. 106, 893 (1957).

[7] J. Friedel, Advances in Physics, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 446.

[8] W. Kohn and S. H. Vosko, Phys. Rev. 119, 912 (1960); N. Bloembergen and T. J. Rowland, Acta Met. 1, 731 (1953).

[9] W. Marshall, Phys. Rev. 110, 1280 (1958).

[10] Y. Koi, A. Tsujimura, T. Hihara, and T. Kushida, J. Phys. Soc. Japan (to be published).