Nuclear Magnetic Resonance of Ni^{61} in Dilute Alloys of Nickel in Cobalt

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The nuclear magnetic resonance of Ni^{61} has been observed in 1% Ni^{61}–99% Co and 2% Ni^{61}–98% Co alloys. The resonance frequency is 70.4 Mc/sec at room temperature and 71.7 Mc/sec at 77°K. The hyperfine field at nickel in cobalt is about two and a half times its value in pure nickel, implying a nearly proportional increase in the local magnetic moment.

I. INTRODUCTION

RECENTLY the measurement of hyperfine fields at the nuclei of ferromagnetic materials and the correlation of these hyperfine fields with the local electronic moments at the atoms have become of increasing interest. Although there are several methods for measuring the product of nuclear moment and hyperfine field at the nucleus, nuclear magnetic resonance is particularly direct, and in the case of nickel well suited, since methods such as the Mössbauer effect, low-temperature specific-heat measurements, γ-ray anisotropy measurements, etc., are either complex or do not have enough sensitivity.

We have recently observed the nuclear magnetic resonance from Ni^{61} in an alloy powder containing 1% nickel and 99% cobalt. A preliminary account of this has been presented.[1] The nickel was isotopically enriched to approximately 100% in the isotope 61. The resonance occurs in zero external magnetic field at 70.4±0.3 Mc/sec at room temperature. We have also observed the Ni^{61} resonance in a 2% alloy with no detectable shift in frequency from the 1% alloy.

II. EXPERIMENTAL PROCEDURES

In preparing the alloy, care was taken to insure that the sample be in the cubic phase and homogeneous. The alloy was melted for 25 min at 1530°C, quenched rapidly, and reheated to 1460°C for 28 h to insure homogenization. The ingot was then ground to less than 10μ particles and annealed for one hour at 500°C. X-ray diffraction showed the alloy to be at least 90% cubic.

About 2 g of the finely divided alloy powder was inserted in the tank circuit of a self-quenched super-regenerative spectrometer of the type used in quadrupole resonance spectroscopy.[2] The nuclear resonance signal was observed directly on an oscilloscope. The signal from Ni^{61} is only about one-fourth as intense as that from pure unenriched[3] nickel metal (the pure nickel resonance occurs at 26.0 Mc/sec at room temperature).

The super-regenerative spectrometer, although very sensitive and well suited for observing nuclear resonance in these materials, has the disadvantage that it does not oscillate at constant amplitude but rather in pulses which, in our experiments, repeat at a rate approximately equal to the nuclear resonance linewidth. The oscillator rf output therefore contains a central frequency ν_0 and equally spaced side bands separated in frequency by the pulse repetition rate (about 250 kc/sec in this experiment). This means that the resonance condition will be satisfied by several values of ν_0 resulting in a "side band pattern" in the observed nuclear absorption. When one attempts to measure the frequency of the oscillator with an auxiliary oscillator, a similar side band pattern occurs; one can easily be in error by about one side band, i.e., the quench frequency, and this represents the chief source or error in the frequency measurements. Measurements of linewidths that are comparable to the quench frequency are subject to large errors.

III. RESULTS

The nuclear resonance has been observed at both room temperature and 77°K in the 1% alloy.

The same nuclear resonance frequency has also been observed in an alloy of 2% Ni^{61} in cobalt. The linewidth was 450 kc/sec±200 kc/sec at room temperature.

IV. DISCUSSION

If we assume that the hyperfine field is approximately proportional to the local magnetic moment[4] at a nickel

Temperature (°K)	Frequency (Mc/sec)	Linewidth (kc/sec)
298	70.4±0.3	300±200
77	71.7±0.3	700±300

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site, as has been found to be true for a given atom in a number of cases, then a hyperfine field for nickel in cobalt of 2.7 times its value in pure nickel implies an increase in the local magnetic moment of the nickel atom from 0.6 Bohr magnetons to 1.6 Bohr magnetons. This argument is independent of the Ni^{61} nuclear moment.

The local moment on the nickel atoms in these cobalt rich alloys may be estimated in another, independent, way. The average local moment $\langle\mu\rangle$ in any cobalt-nickel alloy may be expressed as follows:

$$\langle\mu\rangle=(1-c)\bar{\mu}_{\mathrm{Co}}(c)+c\bar{\mu}_{\mathrm{Ni}}(c),$$

where c is the concentration of nickel and $\bar{\mu}_{\mathrm{Co}}(c)$ and $\bar{\mu}_{\mathrm{Ni}}(c)$ are the average local magnetic moments on the cobalt and nickel atoms, respectively.

The average local moment $\langle\mu\rangle$ is known from saturation magnetization measurements. If it is assumed, as was done by Johnson, Ridout, Cranshaw, and Madsen,[5] that the local moment on the cobalt atoms is proportional to the hyperfine field at the cobalt nuclei then $\bar{\mu}_{\mathrm{Co}}$ may also be evaluated as a function of nickel concentration. The specific heat data of Arp, Edmonds, and Petersen[6] and the nuclear resonance results for nickel rich alloys[7,8] have been used to estimate $\bar{\mu}_{\mathrm{Co}}$. Combining this estimation with the saturation moment data leads to a value of $\bar{\mu}_{\mathrm{Ni}}=1.4$ Bohr magnetons in cobalt rich alloys, that is, when c goes to zero. This is in good agreement with the value of 1.6 Bohr magnetons obtained above, indicating that the local moment is, within 15% or so, proportional to the hyperfine field.

Alternatively, one could assume that the local moment is not appreciably changed despite the large change in the hyperfine field.[9] In this case one might expect that although neighboring cobalt atoms do not change the local moment at the nickel atom appreciably, they do increase the hyperfine field at the nickel atom. We feel that the major change in hyperfine field is due to changes in the local moment.

Bennett and Streever[10] recently proposed a nuclear moment of 0.9 nm for Ni^{61}, rather than the previously estimated value of 0.3 nm.[11] Subsequently, a value higher than 0.3 nm has also been indicated by the shift of the single-domain resonance of Ni^{61} in pure nickel metal.[12] A value of 0.3 nm leads to a field for nickel in cobalt of 462 kOe at room temperature, while a value of 0.9 nm for the nuclear moment puts the hyperfine field at 154 kOe, which is lower than the 217-kOe hyperfine field at cobalt in pure cobalt[13] as expected from the value of the local moment obtained above. The sign of the hyperfine field of nickel in cobalt has not as yet been determined, but since the hyperfine fields are negative in the pure metals (Fe, Co, Ni), the above implies it should be negative for nickel in cobalt also.

The relative intensity of the resonance compared to nickel in pure nickel can be understood as follows: In the absence of saturation effects, the nuclear signal for the same nucleus is expected to be proportional to $(\nu_0/\Delta\nu)N$. Here N is the number of resonating nuclei, and $\Delta\nu$ is the linewidth. The temperature, applied rf field, Q of the coil, and filling factor are all assumed to be constant. Since Ni^{61} occurs naturally with an abundance of 1.25%, the use of 100% Ni^{61} in making the 1% $Ni^{61}-99\%$ Co alloy results in a comparable value of N between the pure nickel and the alloy. The net result should be reduction in signal intensity by a factor of about 7 due to the increased linewidth of the Ni^{61} resonance from the alloy over that in pure nickel (the Ni^{61} linewidth in pure nickel is about 40 kc/sec.) This compares favorably with the observed reduction.

The temperature dependence of the resonance is expected to follow approximately the temperature dependence of Co^{59} in pure cobalt since both resonance frequencies are nearly proportional to the saturation magnetization of the host. The ratio of the Ni^{61} frequency in cobalt at 77°K to its value at 298°K is $71.7/70.4=1.018\pm0.009$; the corresponding ratio for Co^{59} in pure cobalt[13] is $216.9/213.1=1.018$, in good agreement.

The relatively small increase in resonance linewidth in the 2% alloy is interesting since the linewidth of Co^{59} in an alloy of 1% nickel in cobalt is also found to broaden only slightly between 1% and 2%.[14,15] The change in linewidth of the impurity and host resonances with concentration has been briefly discussed previously,[16] with particular reference to dilute alloys of cobalt in nickel. More accurate linewidth data will be necessary to fully understand the origin of the linewidth in ferromagnetic materials.

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