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NMR Studies of Dilute Alloys of Ni in Cu[†]

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We have studied the nuclear magnetic resonance of Cu atoms which are near neighbors to Ni atoms in dilute Cu Ni alloys. The experiments were performed at liquid-helium temperatures and at magnetic fields from 6 to 60 kG. The resonances show up as weak satellites to the main absorption of more distant Cu. By varying the magnetic field, we can show that several lines arise from a single shell of neighbors having a quadrupole coupling $\nu_q = 1.1 \pm 0.1$ MHz, asymmetry parameter $\eta = 0.20 \pm 0.05$, and a magnetic shift $\Delta H/H = -0.27 \times 10^{-3}$. Comparing the quadrupole coupling with estimates made by Béal-Monod and Tompa for CuNi by measuring wipe-out number, we conclude the lines arise from first neighbors. The magnetic shift is smaller than we had previously observed for CuCo in almost exactly the ratio of the χJ 's of the two impurities, where χ is the impurity susceptibility and J is the s - d exchange coupling.

I. INTRODUCTION

Over the past several years there has been extensive interest in the occurrence or lack of occurrence of magnetization when transition-metal atoms are present at low concentration in nonmagnetic hosts such as copper or gold. These are systems in which the Kondo effect is often observed. For the Kondo effect it is thought that at low tempera-

tures the conduction electrons pair up with the spin of the impurities to form a many-body singlet. One of the most powerful techniques for study of these systems has been nuclear magnetic resonance. Heeger and his collaborators through a series of papers¹ have studied the broadening of the resonance of the host copper produced by the magnetic atoms especially for the system copper containing iron. Potts and Welsh² have extended these data.

Allou³ has observed the resonance due to aluminum atoms which are near neighbors to the Mn atoms present in Al. Chapman and Seymour⁴ have observed the wipe out of intensities of the resonance of the copper main line in the system CuNi; Lumpkin⁵ has done the same for CuMn. Narath and Gossard⁶ have studied the V and Ag resonances in Au(Ag)V and Narath⁷ has observed resonances from the magnetic atom itself in a number of systems.

We have observed the weak resonances, satellites, from copper atoms which are near neighbors to Co and Ni in dilute alloys of these atoms. We have looked unsuccessfully in dilute alloys of Cu containing Fe. In this paper we report our results on CuNi. A preliminary account of our work on CuCo has already appeared.⁸

The satellites were first discovered by one of us (D. C. L.) using an apparatus that operated at fields below 10 kG. D. V. L. extended the results to 60 kG using a different apparatus. We present the results of both on CuNi in this paper, as well as a brief description of the low-field rig. The high-field equipment will be described in a later paper on CuCo. Experiments on CuNi were performed at 1.5 and 4.2 K. At 77 K no lines were observed using the low-field equipment.

The samples were prepared by one of us (J. B. B.) assisted later by Stakelon. We have used concentrations of 0.54- and 0.71-at. % Ni. It is known that at these concentrations magnetic susceptibility experiments are strongly disturbed by the presence of pairs or higher clusters of the impurities. One of the desirable features of nuclear-magnetic-resonance studies of satellites is that one can avoid these effects for two reasons. The first is that since the resonance is a spectroscopic method, lines of different species can exist together and be identified as being from different species through the concentration dependence of their intensities. The second reason is that statistically pair or higher spectra are smeared in frequency since there are so many ways of forming clusters of a given size. We have in fact found no lines which we can attribute to such clusters.

In Sec. II we discuss the experimental method. In Sec. III we present the experimental results. The results are summarized in Sec. IV.

II. EXPERIMENTAL METHOD

A. Apparatus

The nuclear-magnetic-resonance spectrometer used in the experiments below 10 kG is a steady-state single-coil rig which has a hybrid tee as the power-splitting element. A block diagram is shown in Fig. 1. The rf field is generated either by a crystal-controlled oscillator with buffer amplifier in its last stage to match its output imped-

ance to 50 Ω or by a commercial frequency synthesizer. The sinusoidal rf magnetic field in the sample coil is adjusted by a 0-120-dB attenuator manufactured by Hewlett-Packard (Models 355C and 355D). When the rf power reaches the port A of the hybrid tee it divides equally between the output ports C and D if their terminations are matched in impedance. When thus matched, the rf level at port B is a minimum, 60 dB below port A. The matching of the sample circuit to 100 Ω is accomplished by the capacitor divider C_1 and C_2 in Fig. 1. The nuclear-resonance-detection part of the circuit is analogous to a conventional bridge-balance rig. The absorptive and dispersive parts of the signal are chosen by adjusting the rf phase shifter. The field modulation was chosen at 150 Hz to minimize noise. The lock-in amplifier was a Princeton Applied Research Model HR-8. A Northern Scientific NS550 signal averager was used to improve the signal-to-noise ratio.

Since the experiment involves a slow sweep at moderate rf power, stability of the rf balance is of great importance. We have taken great care in designing and construction of the rig to cut down the noise due to electromagnetic interaction and mechanical vibration. A great improvement in signal-to-noise ratio was achieved by changing C_1

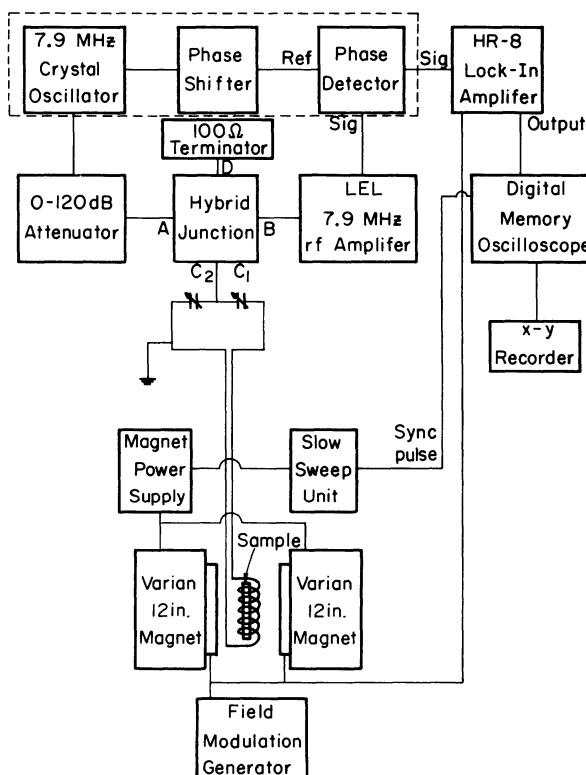


FIG. 1. Block diagram of the apparatus of D. C. L. used below 10 kG.

and C_2 from two standard unshielded butterfly variable capacitors to two General Radio type-1422 precision capacitors which are of rigid construction, have well-shielded cases, and are more stable when the room temperature changes slightly. We have also used GR 50- Ω coaxial rigid air lines to connect the hybrid tee to the two capacitors as well as from the capacitor to the connector in the top plate of the probe to achieve higher mechanical stability.

The sensitivity of the hybrid junction spectrometer is such that for most of our samples (except CuFe, which has a severe magnetic broadening) the signal-to-noise ratio of the main copper line at 1.4 K and 7 kG is better than 3×10^3 with a 4-G peak-to-peak modulation and 3-sec lock-in RC-time-constant setting.

B. Samples

The alloys were normally prepared by melting 99.999%-purity copper metal with impurity metal (with 99.999% purity) in appropriate concentration ratios in an argon atmosphere and kept in an induction furnace (1200–1260°C) for 1 h. Then the melt was quenched rapidly in cold water. A swaging process followed, reducing the ingot diameter by 40–50%. The swaged ingot was then annealed at 1010 to 1060°C for 3–4 days in a quartz crucible filled with argon gas. Then the annealed ingot was quenched rapidly in ice water to avoid precipitation. The samples were converted to powder by a tungsten-carbide rotary cutter and passed through a 400-mesh sieve (i. e., with diameter less than 37μ). Some of the powder was annealed to reduce the dislocations introduced by filing. Before this annealing the filings were pickled in HCl, rinsed in deionized water, dried, and then mixed with Al_2O_3 powder. The powder was annealed in an Ar-filled quartz ampule at 1010–1060°C for 2 h and then quenched rapidly in ice water. The strain-relieving annealing process eliminates some quadrupole wipe out. Microprobe analysis of the more concentrated samples showed the alloys to be homogeneous to within about $\pm 5\%$. A multielement mass-spectrographic analysis showed that extraneous impurity contamination was minimal. Chemical analysis determined the Ni concentration.

C. Experimental Procedures

Nuclear-magnetic-resonance measurements were made at frequencies between 6.4 and 9.2 MHz with the hybrid-tee spectrometer at 1.4, 4.2, and 77 K and up to 60 MHz at 1.4 and 4.2 K using the second spectrometer. The sample is always in contact with the liquid helium or the liquid nitrogen when the experiment is run at these temperatures. The 150-Hz field modulation had a peak-to-peak

amplitude normally of 4 G, about as high a value as could be used without unacceptable distortion of the signal.

The radio-frequency exciting field was kept at about 3 dB below the value required to give an obvious saturation effect. A home-built improved Pound box was used for magnetic field strength measurement.

An important method for improving detection of satellites close to the main line was to readjust the rf reference phase to give that mixture of χ' and χ'' as to provide a flat base line. This effect is shown in Fig. 2 which displays a variety of phase settings for Cu-0.7-at.% Ni at 51.5 MHz and 4.2 K. Note the difficulty in detecting the satellite (satellite M) under pure absorption, and how strongly it is resolved under a mixture. Note also that the peak position one would deduce by eye does not change radically with phase. The range of the sweeping magnetic field was about 80 G, the time per sweep was about 210 sec., and the lock-in amplifier time constant was set at 1–3 sec.

III. EXPERIMENTAL RESULTS

A. Review of Theory and Previous Experiments

The magnetic impurity shifts the resonance of the neighboring copper nuclei. We distinguish

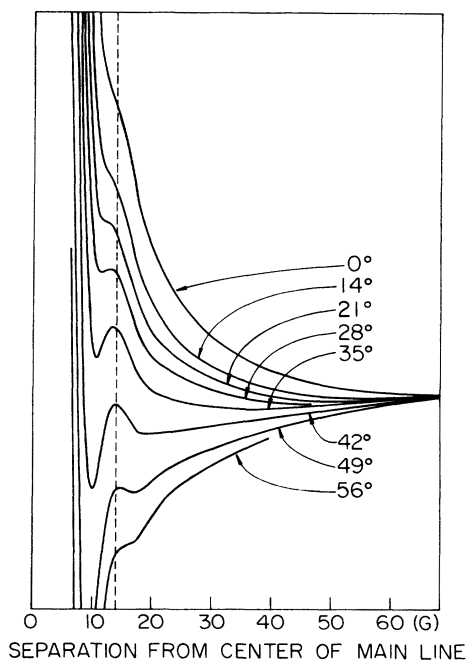


FIG. 2. Effect of adjusting the phase θ of the rf reference signal to vary the mixture of χ' and χ'' on one's ability to perceive the satellite. The vertical axis is proportional to a linear combination of the derivatives with respect to magnetic field H of the absorption and dispersion: $(\partial\chi'/\partial H) \sin\theta + (\partial\chi''/\partial H) \cos\theta$.

TABLE I. Field gradients at various shells (after Béal-Monod, Ref. 12).

Neighbor	First	Second	Third	Fourth
ν_q (MHz)	1.39	0.525	0.156	0.104
Nuclei in shell	12	6	24	12

three aspects.

(a) The magnetic moment of the impurity both directly and indirectly through polarizing the conduction electrons produces a magnetic shift of the neighbors ΔH_m . This shift depends on the crystallographic location of the neighbor relative to the impurity (for example to which shell of neighbors the Cu belongs) and is proportional to the applied field:

$$\Delta H_m = AH . \quad (1)$$

(b) Since Cu has spin $\frac{3}{2}$, there is a quadrupole splitting in first order of both the $\frac{3}{2}$ to $\frac{1}{2}$ and the $-\frac{3}{2}$ to $-\frac{1}{2}$ transitions. The first-order quadrupole splitting is independent of the applied field. Moreover, for every positive splitting there is a corresponding negative one:

$$\Delta H = +B \text{ and } \Delta H = -B . \quad (2)$$

(c) The $\frac{1}{2}$ to $-\frac{1}{2}$ transition is not split by a quadrupole coupling in first order, but is to second order. Therefore, this splitting is inversely proportional to the field

$$\Delta H = C/H . \quad (3)$$

Since calculation of the splitting frequencies depends on the orientation of the crystal with respect to the applied field, and since powder samples are used, the theoretical spectrum requires a powder average.

Drain⁹ and Baugher, Kriz, Taylor, and Bray¹⁰ reviewed the form of the powder pattern for first- and second-order quadrupole couplings, respectively.

The general effect of magnetic and quadrupole coupling between the magnetic atoms and the neighboring Cu nuclei will produce NMR signals that fall into three classes: (i) lines with sharp features, well split from the main resonance; (ii) well-split lines not exhibiting features, i. e., a general smear; (iii) lines not split from the main line.

Unless one is looking with excellent sensitivity, the quadrupole effects (b) and (c) simply remove intensity from the main line. One then talks about "wipe out," in fact first-order quadrupole wipe out and second-order quadrupole wipe out going with cases (b) and (c), respectively.¹¹

Using this method Béal-Monod¹² and Tompa¹³ have studied CuNi. They obtain phase shifts for $l=0, 1,$ and 2 scattered waves. Béal-Monod's estimates of the quadrupole frequency are given in Table I. These values are based on the application of the Kohn-Vosko¹⁴ and Blandin-Friedel¹⁵ theory. However, since that theory is only approximate for the first few neighbors, one should not place too great a reliance on the precise numerical value.

B. Experimental Results

In our work we focus on resolved features. We use the magnetic field dependence to determine the magnetic coupling and to determine whether or not we are seeing case (b) or (c) as far as quadrupole effects are concerned.

Figure 3 shows the CuNi resonance at 7.0 kG. Several peaks are clearly seen (labeled A, B, C, and M and N).

To distinguish the various quadrupole cases (b) and (c) we expect a line with a first-order splitting to go as

$$\Delta H = A H + B , \quad (4)$$

or if there is a second-order splitting,

$$\Delta H = A H + C/H . \quad (5)$$

Figure 4 shows a plot of the five peaks versus $1/H$. We see that the three peaks B, C, and N are straight lines for low to moderate field strengths (right side of the figure), but evidently coalesce to a single line at high field. They therefore represent features of a second-order quadrupole shift of a $\frac{1}{2}$ to $-\frac{1}{2}$ transition as given by Eq. (5). Comparison with the paper by Baugher *et al.*¹⁰ shows that these peaks look a great deal like their Fig. 4. Peaks B and C would be a single peak if the field gradient were axially symmetric. In addition the ratio of their splitting to that of peak N would be 16/9. Figure 5 shows a computer-generated theoretical powder pattern fit to peaks B, C, and N calculated on a $\Sigma 5$ computer with the assistance of Thomas Aton showing how the pattern changes with changing η . The theoretical fit corresponds to $H_0 = 7$ kG. The best fit to the data gives $\nu_q = 1.1 \pm 0.1$ MHz, $\eta = 0.20 \pm 0.05$.

Examination of Table I shows that this ν_q goes with either the first or second neighbor to the impurity. We can rule out the second-neighbor possibility, however, since its fourfold symmetry axis guarantees that it have $\eta = 0$, for which case the peaks B and C would not be split. Thus we assign these lines to the first neighbor.

Peaks A and M are independent of field. Since they are symmetrically displaced from the main line, they represent first-order splittings with negligible magnetic splitting such as are given by Eq. (4). In our first work at low fields, it was not

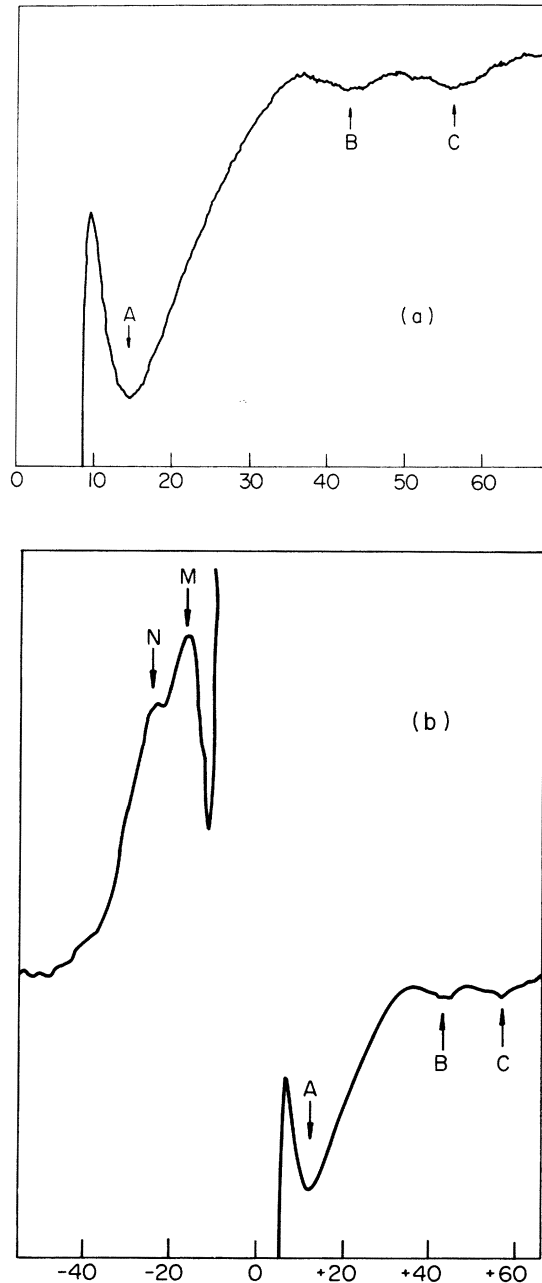


FIG. 3. Derivative of absorption versus displacement of magnetic field in gauss from main Cu⁶³ resonance. (a) Satellites on the high-field side of the main Cu resonance in Cu - 0.71-at.% Ni. ($H_0 = 7$ kG, average of 20 sweeps, lock-in time constant 3 sec.) (b) Sweep showing both high- and low-field sides displaying relative peak heights on the two sides, and labeling the peaks A, B, C, M, and N for reference to the text.

possible to decide whether peaks A and M did or did not move with the field. If we assumed they also obeyed Eq. (5), we could explain the spectrum as a mixture of two second-order spectra superposed.

The data of Fig. 4, however, show clearly that A and M do not move with field so that we can rule out the possibility of two second-order patterns.

If peaks A and M represent the powder pattern of a first-order quadrupole interaction with $\eta = 0$, ν_q will be either 28 or 14 kHz depending on whether they correspond to logarithmic singularities or step singularities, respectively, in the powder pattern. We do not have enough information, however, to tell whether or not $\eta = 0$. In fact we do not know whether or not these peaks represent more than a single neighbor shell.

Further comparison with Table I shows that if peaks B, C, and N come from the first neighbor, the $\frac{1}{2}$ to $-\frac{1}{2}$ shift of the further shells will be too small for them to be resolved from the main line, so that Table I is consistent with our failure to observe other second-order lines.

On the other hand, the first-order splittings of the $\frac{3}{2}$ to $\frac{1}{2}$ and $-\frac{1}{2}$ to $-\frac{3}{2}$ transitions are probably too broad to detect for these first neighbors, leading to our observing only the general humps of peaks A and M.

Schumacher and Schnakenberg¹⁶ have done single-crystal studies on CuZn, the alloy which is on the opposite side of pure Cu from CuNi in the Periodic Table. They find a first neighbor $\nu_q = 1.960$ MHz and an $\eta = 0.224$, comparable to the CuNi first-neighbor coupling.

The magnetic shift is $\Delta H/H = -(0.27 \pm 0.03) \times 10^{-3}$. Table II gives the measured magnetic shift for the CuNi first neighbor together with pertinent data for CuCo.⁸ The experimental susceptibility contains both spin and orbital contributions. The values of the exchange coupling J are gotten from the Kondo temperatures using the formula¹

$$T_K = T_F e^{-1/2 \rho |J|}, \quad (6)$$

where ρ is the density of states at the Fermi energy (0.15 eV/atom for copper) and T_F is the Fermi energy (8.2×10^4 K).

TABLE II. Comparison of CuCo with CuNi alloys.

X	CuCo	X_{CuCo}/X_{CuNi}	CuNi
T_K (K)	4900 ^a		6900 ^b
J (eV)	1.19	0.88	1.35
χ (emu/atom)	4.0×10^{-27} c	16.7	0.24×10^{-27} b
$J\chi$	4.76×10^{-27}	14.7	0.324×10^{-27}
$\Delta H/H$	-3.84×10^{-3} d	14.2	-0.27×10^{-3}

^aFrom the temperature dependence of $\Delta H/H$ of the first shell of neighbors CuCo, Ref. 7.

^bE. W. Pugh, B. R. Coles, A. Arrott, and J. E. Goldman, Phys. Rev. **105**, 814 (1957).

^cR. Tournier and A. Blandin, Phys. Rev. Lett. **24**, 397 (1970).

^dFrom Ref. 8.

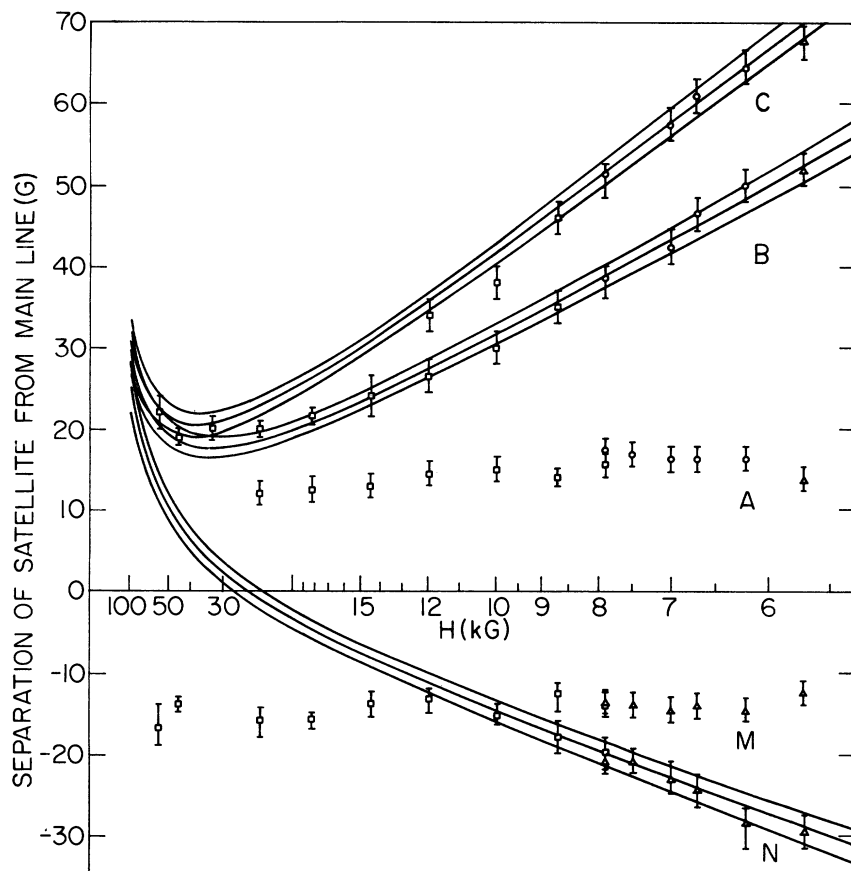


FIG. 4. Peak positions vs $1/H$ showing three field-dependent lines and two field-independent lines. The theoretical curves are computed assuming the field-dependent lines are all features on the $+\frac{1}{2}$ to $-\frac{1}{2}$ transition of a line possessing a magnetic shift and a second-order quadrupole coupling. The three families of trios of closely spaced theoretical lines correspond to $\Delta H = 0.30H_0 + (C+10)/H_0$, $\Delta H = 0.27H_0 + C/H_0$, $\Delta H = 0.24H_0 + (C-10)/H_0$, with H_0 in kG and $C = 390, 290, -175$.

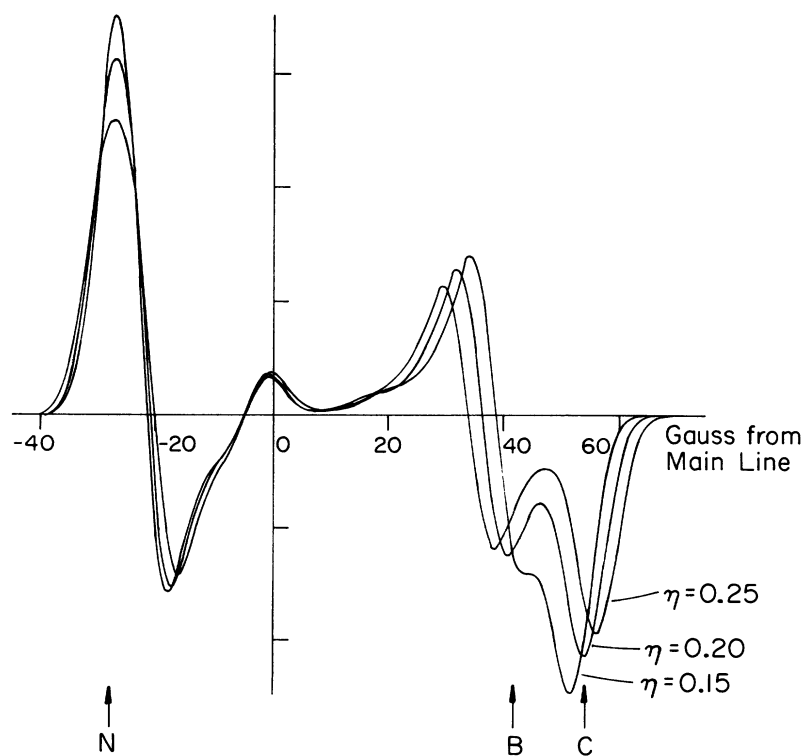


FIG. 5. Theoretical powder pattern for a $+\frac{1}{2}$ to $-\frac{1}{2}$ transition at $H_0 = 7$ kG illustrating how the variation of η causes the right-hand peak to split. The split peak we associate with experimental lines B and C, and the large peak on the opposite side with experimental peak N.

The strength of the magnetic shift $\Delta H/H$ of the first neighbor should be, in the Rudermann-Kittel-Kasuya-Yosida (RKKY) approximation,¹ proportional to χJ . We note from Table II that the relative shifts for CuCo to CuNi is well described by this factor, though the ratio of J 's is so close to unity that the ratio of χ 's alone accounts for most of the shift. In view of the mixture of orbital and spin contributions to χ we can at most conclude, therefore, that the relative shifts are quite reasonable. Of course as Geldart¹⁷ has emphasized, we do not expect the RKKY expression to be accurate at the first neighbor and in fact for both CuCo and CuNi RKKY predicts a $\Delta H/H$ of the wrong sign.

IV. CONCLUSIONS

We have resolved several satellites to the main line of Cu in CuNi alloys. From the field dependence and the wipe-out studies of Béal-Monod and Tompa we can identify one set as arising from the first shell of neighbors. That shell has a quadrupole coupling ν_q and asymmetry parameter η which are quite similar to the first neighbors to Zn in

CuZn, a reasonable result since Ni and Zn are the two neighbors of Cu in the Periodic Table.

The magnetic shift of the first neighbor $\Delta H/H = -0.27 \times 10^{-3}$ is an order of magnitude smaller than for CuCo, but almost exactly scales in the ratio χJ , where χ is the susceptibility of the impurity, and J the exchange coupling in the s - d Hamiltonian, as would be expected from simple theories, such as RKKY. The sign of the coupling, however, is wrong, as has been observed by Geldart¹⁷ previously. Since the ratio of the J 's of CuCo to CuNi is close to unity, and since the χ 's contain contributions from both spin and orbit, the shift could scale with χ only.

Nothing about the CuNi data would require the full theoretical framework of the usual Kondo description.

ACKNOWLEDGMENTS

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