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Nuclear Magnetic Relaxation of the Impurity Nucleus in Dilute Ferromagnetic Alloys

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We have measured the nuclear relaxation times T_1 and T_2 for 1% Ni^{61} in 99% iron, 1% Ni^{61} in 99% cobalt, and 0.5% Co^{59} in 99.5% nickel at several temperatures using the pulsed-free precession method. The relaxation curves are generally nonexponential and power-dependent. The signals at low-power levels are from nuclei in domain walls and T_1 is due to thermal fluctuations of the domain walls. At high-power levels, where the signal is mainly from nuclei in domains, the longest measured relaxation times T_1 are lower limits for relaxation times for nuclei in domains. The longest T_1's are found to be inversely proportional to temperature with $T_1T=1.0$ sec °K for 1% Ni^{61} in cobalt, $T_1T=1.2$ sec °K for 1% Ni^{61} in iron, and $T_1T=0.3$ sec °K for 0.5% Co^{59} in nickel, and are believed to be due to conduction electron relaxation. The quantity $1/\gamma_n{}^2T_1T$ (where γ_n is the nuclear gyromagnetic ratio) is a normalized measure of the strength of the conduction electron relaxation mechanism and is found to be smaller for the impurity nuclei than for the nuclei of the solvent atoms in the pure solvent metal. This is believed to show that the relaxation of the nuclei by the conduction electrons (which is enhanced in the pure metals by spin waves) is slower for the nuclei on the solute atoms in the alloys due to a reduction of the spin wave enhancement. T_2 for Ni^{61} in cobalt is 0.15 msec at room temperature and is believed to be due to a coupling of Ni^{61} and Co^{59} nuclei by the Ruderman-Kittel interaction.

I. INTRODUCTION

IN the pure metals, iron, cobalt, and nickel, the nuclear relaxation times T_1 and T_2 have been measured by Weger[1–3] by the free precession method. The decay of the longitudinal and transverse magnetization in the ferromagnetic metals is found to be nonexponential and a function of the rf power level. Nuclei in the domain walls experience a large enhancement of the applied rf level due to the domain wall motion. Thermal fluctuations of the domain walls also provide a relaxation mechanism; consequently, at low rf pulse levels, the nuclear signals are from nuclei near the center of the domain walls which have the largest rf enhancement and shortest relaxation times. At higher rf pulse amplitudes signals come from nuclei farther out from the centers of domain walls, which have lower enhancements of rf fields and longer relaxation times. Finally, at very high rf pulse levels, signals come primarily from nuclei in the domains which see a much smaller rf enhancement than those in the domain walls. For these nuclei, relaxation times T_1 are longer, being determined by conduction electrons now and no longer by domain wall motions. For T_2, the experimental situation is more complicated. For low abundance of nuclear spins and at high temperatures the nuclear spin-spin interactions can generally be neglected. In this case, T_2 is found to be of the same order as T_1. For cases in which nuclear spin-spin interactions are important, as in pure cobalt, these determine T_2.

In the present work we have used the free precession method to measure T_1 and T_2 for Ni^{61} in dilute alloys of 1% Ni^{61} in cobalt[4] and iron,[5] and for Co^{59} in a 0.5% Co^{59} in nickel alloy.[6] Preliminary to studying relaxation times of the impurity nuclei in the alloys, some measurements were also made of T_1 and T_2 for Ni^{61} in pure unenriched nickel powders to determine the effect of annealing on the relaxation times. The Ni^{61} relaxation times were also measured in the 0.5% cobalt in nickel alloy to determine the effect of the cobalt impurity on the Ni^{61} relaxation.

In Sec. II the experimental methods and techniques are discussed. In Sec. III we review briefly the theory of relaxation, both in domain walls and in the domains. In Sec. IV we give the experimental results, and compare them with the theory.

II. EXPERIMENTAL METHOD AND TECHNIQUE

A. Equipment

We have used the free precession method to measure the inhomogeneous linewidth and the longitudinal and transverse relaxation times T_1 and T_2. A pulsed push-pull rf oscillator (Arenberg Ultrasonic Laboratories Model 650-C) was used to supply the rf pulses. The widths of the rf pulses used were about 1 μsec and the maximum rf voltage level was about 300 V peak-to-peak rf. The metal powder was inserted in the rf coil of the oscillator. A second coil coaxial to the first was used to detect the free precession signal. It was convenient to heterodyne the nuclear signal with a mixing frequency differing by about 20 Mc/sec, using a television tuner. The television tuner was followed by a wide-band rf

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[1] M. Weger, thesis, University of California, Berkeley, 1961 (unpublished).

[2] M. Weger, E. L. Hahn, and A. M. Portis, J. Appl. Phys. 32, 1645 (1961).

[3] M. Weger, Phys. Rev. 128, 1505 (1962).

[4] R. L. Streever, L. H. Bennett, R. C. La Force, and G. F. Day, Phys. Rev. 128, 1632 (1962).

[5] R. L. Streever, L. H. Bennett, R. C. La Force, and G. F. Day, J. Appl. Phys. 34, 1050S (1963).

[6] L. H. Bennett and R. L. Streever, J. Appl. Phys. 33, 1093S (1962).

amplifier and detector. The video signal was then amplified and observed directly on the oscilloscope. The dc pulses necessary to gate the pulsed oscillator were supplied by appropriate combinations of Tektronix Nos. 162 and 163 units.

B. Measuring Methods

To measure T_1 and T_2, one usually employs a method in which one makes use of 90° and 180° pulses. In a ferromagnetic metal as a result of the skin depth and the distribution in enhancement factors, different nuclei see different rf fields. Consequently, 90° and 180° pulses are impossible to obtain. The decay of the magnetization in the transverse direction with time constant T_2 is, however, independent of pulse adjustments and T_2 can be obtained from the echo decay envelope. To measure T_1, we saturated the nuclear signal with one or more closely spaced rf pulses, and then monitored the recovery of the magnetization by observation of the echo following two closely spaced pulses applied some time after the saturating pulses. An echo rather than a single pulse was used to sample the recovery of the magnetization because for the broad resonance lines where the echo width is a few μsec, the receiver blocking after the rf pulse makes the decay itself impossible to observe. Weger[1] has shown that the above method is subject to some error due to diffusion of magnetization in space out of the domain walls. By applying a large number of saturating pulses whose separation is much less than T_2, this effect can to a large extent be overcome.

The relaxation times T_1 and T_2 were measured as a function of rf pulse amplitude. The echo signal following two pulses of equal width goes from zero through a maximum at low rf pulse amplitudes as the pulse amplitude is increased from zero, then slowly decreases again as the pulse amplitude is further increased, as discussed in Ref. 1. The variation in the echo signal intensity results from the distribution in rf fields due to skin depth and a distribution in enhancement factors, and the signal reaches a maximum when $\gamma_n\eta_1 H_1 t_w \sim \pi/2$. Here η_1 is an *average effective* enhancement factor, $2H_1$ is the linearly applied rf field, t_w is the pulse width, and γ_n is the nuclear gyromagnetic ratio. The relaxation curves were in general nonexponential, and a function of the amplitude of the rf pulses for a given pulse width, which was usually about one μsec. T_2 and T_1 can in the case of a nonexponential decay be defined for any point on the relaxation curves from the slope. That is,

$$1/T_2 = -d\ \ln M(t)/dt,$$

where $M(t)$ is the amplitude of the echo signal at a time t after the first pulse and

$$1/T_1 = -d\ \ln[M(\infty) - M(t)]/dt,$$

where $M(t)$ is the value of the sampling signal at a time t after the last saturating pulse.

C. Samples

The majority of the relaxation time studies were made on alloy powders of 1% Ni^{61} and 99% cobalt,[4] 1% Ni^{61} and 99% iron,[5] and 0.5% Co^{59} and 99.5% nickel[6] (all atomic percents). The Ni^{61} in cobalt was annealed for $\frac{1}{2}$ h at 600°C. The Ni^{61} in iron sample was annealed for $\frac{1}{2}$ h at 680°C. The Co^{59} in nickel sample was annealed for 1 h at 1100°C, but some measurements were also made on an unannealed sample. The relaxation times for nuclei in the domain walls is expected to depend somewhat on the degree of particle annealing and will be discussed in Sec. III.A. The average particle size was about 10 μ. More details of sample preparation have been previously discussed.[4–6]

Some studies were also made on the Ni^{61} resonance in pure unenriched nickel samples with various degrees of annealing to investigate the effects of strains on the relaxation times. These samples were all 10 μ powders. The preparation of the pure nickel samples has been discussed previously.[7]

III. THEORY

A. Relaxation in the Domain Walls

Nuclear relaxation in the domain walls has been studied both theoretically and experimentally by several authors.[1,3,8–10] For the pure metals Weger[3] gives for $1/T_1$

$$1/T_1 = (kT\tau_c\omega^2/\delta^2\pi M_s^2)(1+\omega^2\tau_c^2)^{-1}. \tag{1}$$

Here k = Boltzmann constant, T = absolute temperature, τ_c = correlation time for wall motion due to thermal fluctuations, δ = wall thickness, M_s = saturation magnetization, and ω = resonance frequency. For cobalt at room temperature Eq. (1) gives good agreement with experiment.[3] For the pure metals and the alloys studied in this paper, $\omega\tau_c < 1$ is a good approximation. In this limit

$$1/T_1 = kT\tau_c\omega^2/\pi\delta^2 M_s^2. \tag{2}$$

The weak temperature dependence of T_1 in the pure metals is believed to arise because while kT enters directly into Eq. (2), τ_c probably increases with decreasing temperature due to greater eddy current damping of the domain wall motion.[3]

Equation (2) was derived for the pure metals; however, quite generally in the limit $\omega\tau < 1$ the relaxation time will be given by (see Ref. 11)

$$1/T_1 = \langle H_f^2 \rangle \gamma_n^2 \tau_c. \tag{3}$$

Here H_f is the fluctuating field perpendicular to the hyperfine field H_n resulting from thermal fluctuations

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[7] R. L. Streever and L. H. Bennett, Phys. Rev. **131**, 2000 (1963).

[8] J. M. Winter, Phys. Rev. **124**, 452 (1961).

[9] E. Simanek and Z. Sroubek, Czech. J. Phys. **311**, 764 (1961).

[10] M. Matsuura *et al.*, J. Phys. Soc. Japan **17**, 1147 (1962).

[11] C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963), Chap. 5, p. 153.

of the domain wall. Equation (3) can be rewritten

$$1/T_1=\langle\theta^2\rangle\gamma_n{}^2H_n{}^2\tau_c=\langle\theta^2\rangle\tau_c\omega^2\,, \tag{4}$$

where θ is the angular displacement of the electron spin in the domain wall.

At a given temperature Eq. (4) shows that either an increase in the amplitude of the wall motion or the correlation time of the motion will shorten T_1. These parameters should be sensitive to the degree of anneal of the metal. Hence, in the pure metals, for example, in pure nickel, we might expect the Ni^{61} relaxation times T_1 to depend on the degree of annealing of the sample. Also, for the same annealing time we might expect the parameters $\langle\theta^2\rangle$ and τ_c to depend on the concentration of impurities so that the relaxation times T_1 of the solvent nuclei might be expected to depend on the impurity concentration. We also see from Eq. (4) that samples with the highest rf enhancements (due to domain wall motion) will be expected to have the shortest relaxation times for the nuclei in domain walls, a result previously noted.[10]

Consider now the relaxation time of the nuclear spin of the solute atom. From Eq. (4) we see that if the impurity spin in the domain wall moves with the same correlation time and angular displacement as the solvent atom spin in the dilute alloy, then the value of $T_1\omega^2$ for the nuclei of the solute and solvent atoms should be equal. This relation is used in Sec. IV.C to discuss values of T_1 measured for the nuclei of the solute atoms in the domain walls.

B. Relaxation in the Domains

The spin-lattice relaxation time for nuclei in the domains has been considered by Weger.[3] The relaxation time T_1 for nuclei in domains due to conduction electron relaxation is given by the expression

$$1/T_1=(kT\omega^2a^2\Sigma)(h\omega_d{}^2 32\pi^3S^2)^{-1}\,, \tag{5}$$

where a = lattice constant, ω_s = parameter describing spin-wave spectra, S = average spin/atom, Σ = area of Fermi surface, ω = nuclear resonance frequency, k = Boltzmann constant, and T = absolute temperature.

Equation (5) applies to the pure metals and the T_1 mechanism is a second-order process which involves the coupling of the nuclei to the unpaired electron spins through the hyperfine interaction ($A\mathbf{I}\cdot\mathbf{S}$) and the coupling of the electron spins to the conduction electrons through the *sd* coupling. The intermediate states involve the excitation of spin waves.

In the cases where hyperfine fields are proportional to local atomic moments (this is, at least, roughly true for the cases of concern here),[5] the factor (ω^2/S^2) appearing in Eq. (5) is just proportional to $\gamma_n{}^2$, where γ_n is the nuclear gyromagnetic ratio. Quite generally, the quantity $1/\gamma_n{}^2T_1T$ is a normalized measure of the strength of the conduction electron relaxation mechanism. If the mechanism in which the nuclear relaxation is enhanced by the spin waves applies to the solute nuclei in the dilute alloys, as well as to the pure metals, then the quantity $\gamma_n{}^2T_1T$ would be expected to be of the same order of magnitude for the nucleus of the solute atom in the dilute alloy as for the nuclei of the solvent atoms in the same alloy or in the pure solvent metal. One might expect that the creation of spin waves by a spin flip of the solute atom will in general involve a higher energy than in the case of the same process in the pure metal and that the enhanced conduction electron relaxation mechanism will be weaker for the solute nuclei. In this case the quantity $\gamma_n{}^2T_1T$ will be longer for the solute nuclei and closer to its value in the nonferromagnetic materials.

C. Transverse Relaxation

Weger has measured T_2 as well as T_1 in the pure metals; iron, cobalt, and nickel.[1–3] In the case of Ni^{61} or Fe^{57} in the pure unenriched metals, where nuclear spin-spin interactions are weak, T_2, at least at higher temperatures and in the domain walls, is of the order of T_1 and the transverse relaxation is related to the T_1 mechanism. In the case of Co^{59} in pure cobalt where the abundance of active nuclei is high, T_2 is determined by nuclear spin-spin interactions.

At low temperatures in the domains where T_1 is long, T_2 may be determined by a mechanism of the type discussed by Moriya.[12] He considers in detail nuclear relaxation in antiferromagnets; however, similar considerations would apply to ferromagnets as well. Fluctuations in S_z can arise from the interference of two spin waves and might be particularly important in the alloys. These fluctuations would be very effective for relaxation since they occur at lower frequencies than the spin waves. Hence, at low temperatures in the domains, we might expect $T_2<T_1$, even in cases where the nuclear spin-spin coupling can be neglected.

IV. RESULTS AND DISCUSSION

A. T_1 and T_2 for Ni^{61} in Pure Nickel and in the Dilute Alloys

Preliminary to studying relaxation times of the solute nuclei in the dilute alloys, measurements were made of T_1 and T_2 for Ni^{61} in pure unenriched nickel powder with a particle size of 10 μ or less to determine the effect of annealing. The measurements were made at 77°K and the initial relaxation rates were measured at low pulse levels to determine T_1 and T_2 appropriate to the nuclei in domain walls. We found for unannealed samples or for samples annealed for 1 h at 300 and 500°C that T_1 was roughly independent of annealing and about 1.2 msec with T_2 about 0.3 msec. In nickel annealed at 1100°C for 1 h, however, T_1 decreased to 0.5 msec and T_2 to 0.06 msec. The Ni^{61} relaxation time

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[12] T. Moriya, Progr. Theoret. Phys. (Kyoto) **16**, 641 (1956).

appropriate to nuclei in domain walls was also measured at 77°K in a sample of 99.5% nickel and 0.5% cobalt, which was annealed at 1100°C for 1 h. T_1 was found to be 1.3 msec, which is about the same as the T_1 in the pure nickel which was not well annealed.

The decrease in T_1 in the well-annealed samples can be explained as follows: In the well-annealed nickel, the initial permeability is high and we expect a larger amplitude for the domain wall motion (for a given excitation). This is in agreement with previous continuous-wave studies,[7] where the large dispersion signals in the well-annealed samples were explained partly by the larger rf enhancements in these samples. The decrease in T_2 is partly related to the T_1 decrease; however, the ratio of T_1 to T_2 is even greater in the well-annealed sample, indicating that fluctuations in S_z along the direction of the nuclear hyperfine field may be more important here. We also see that in the well-annealed sample of 0.5% cobalt in nickel the Ni^{61} relaxation time T_1 is about the same as in the unannealed pure nickel. Both strains as well as impurities seem to have the effect of limiting the displacement of the domain walls and lengthening the relaxation time. For the same annealing time at least the relaxation time is longer in the 0.5% cobalt sample than in the pure nickel.

B. T_1 and T_2 of the Solute Nuclei in the Alloys

The longitudinal and transverse relaxation curves were obtained as a function of pulse level at various temperatures, as discussed in Sec. II.B, for the solute nuclei in annealed samples of 1% Ni^{61} in cobalt, 1% Ni^{61} in iron and 0.5% Co^{59} in nickel. The relaxation curves were, in general, nonexponential and dependent on the pulse level. The longest and shortest times are given in Table I. The shortest times T_{10} and T_{20} were determined from the initial slopes of the respective relaxation curves obtained at a low pulse amplitude below that at which the echo signal went through a maximum, and are believed to approach the shortest relaxation times appropriate to nuclei near the center of the domain walls. The longest times T_{1M} and T_{2M} were measured at the highest pulse level from the slope of the respective relaxation curves at the farthest time out, and are believed to approach the values for nuclei in the domains. The highest rf pulse level which was roughly 12 times that at which the echo signal went through a maximum corresponded to an H_1 of roughly 6 G for Ni^{61} in cobalt and Ni^{61} in iron and about 3 G for Co^{59} in nickel.

TABLE I. Relaxation times for the impurity nuclei in the dilute alloys, T_{10} and T_{20} are the relaxation times appropriate to nuclei in domain walls, while T_{1M} and T_{2M} approach the relaxation times in domains as discussed in the text.

		1% Ni^{61} in Cobalt		
Temp. (°K)	T_{10} (msec)	T_{1M} (msec)	T_{20} (msec)	T_{2M} (msec)
300	1.4	3.5	0.15	0.15
77	2.0	13	0.18	0.18
4	3.5	250	0.19	0.22
		1% Ni^{61} in Iron		
Temp. (°K)	T_{10} (msec)	T_{1M} (msec)	T_{20} (msec)	T_{2M} (msec)
300	0.7	4	0.17	2.0
77	1.7	15	0.6	2.4
4	6.0		1.0	3.0
		0.5% Co^{59} in Nickel		
Temp. (°K)	T_{10} (msec)	T_{1M} (msec)	T_{20} (msec)	T_{2M} (msec)
300	0.05	1	0.03	0.12
77	0.13	4	0.04	0.18
4	1.4		0.15	0.34

The longitudinal relaxation curves were dependent on the rf pulse level, nonexponential at low pulse levels and more nearly exponential at higher pulse levels, where the signal was mainly from nuclei in domains. For the case of Ni^{61} in iron and Co^{59} in nickel, the transverse decay curves at the higher temperatures were also dependent on the pulse level and nonexponential and T_2 in the domain walls was of the same order as T_1, indicating it was related to the T_1 mechanism. At lower temperatures, particularly in the domains, T_2 is seen from Table I to be much less than T_1 and less dependent on pulse level. In the case of Ni^{61} in cobalt, the transverse decay curves are nearly exponential and T_2 was nearly independent of pulse level. The nearly unique value of T_2 for this case indicates it is being determined by nuclear spin-spin interactions, a similar result having been observed for the case of Co^{59} in pure cobalt metal.[1]

The inhomogeneous linewidths at room temperature obtained from the width of the echo (assuming an exponential decay) and measured by the quantity $1/\pi T_2^*$ were about 240 kc/sec for 1% Ni^{61} in cobalt and 190 kc/sec for 0.5% Co^{59} in nickel and were in approximate agreement with continuous wave measurements. The 1% Ni^{61} in iron width ($\sim$700 kc/sec) was too broad to estimate accurately from the width of the echo.

C. Discussion of T_1 in Domain Walls

In Table II we have compared the value of T_{10} (the T_1 appropriate to nuclei in domain walls) for the nuclei in the dilute alloys with the shortest T_1's measured in the pure solvent metals by Weger[2] which correspond to our T_{10}'s. In view of Eq. (4), if the angular displacement and the correlation time associated with the thermal fluctuations of the solute electron spin in the domain wall were the same as for the electron spin in the pure solvent metal, then the quantity $T_{10}\nu^2$ (where ν is the resonance frequency) should be the same for the solute nucleus as for the nucleus in the pure solvent metal. In the last two columns of Table II we compare the quantity $\nu^2 T_{10}$ for the solute nuclei in the alloys and the nuclei in the pure solvent metal. We see that in all cases this quantity is longer for the solute nuclei. This can probably be understood as due to the fact that the thermal fluctuations of the domain wall in the dilute

TABLE II. A comparison of the longitudinal relaxation times in the domain walls, T_{10}, for the impurity nuclei in the dilute alloys (from Table I) with the corresponding times in the pure host metal measured by Weger *et al.*[a] The quantities $\nu^2 T_{10}$ are also compared.[b]

T_{10} for Co^{59} in pure cobalt and 1% Ni^{61} in 99% cobalt.

Temp (°K)	T_{10} (msec)[a] Co^{59}	T_{10} (msec) Ni^{61}	ν (Mc/sec) Co^{59}	ν (Mc/sec) Ni^{61}	$\nu^2 T_{10}$ (Mc^2/sec) Co^{59}	$\nu^2 T_{10}$ (Mc/sec) Ni^{61}
300	0.1	1.4	213	70	4.5	6.9
77	0.2	2.0	217	72	9.4	10.4
4	0.2	3.5	217	72	9.4	18.1

T_{10} for Fe^{57} in pure iron and 1% Ni^{61} in 99% iron.

Temp (°K)	T_{10} (msec)[a] Fe^{57}	T_{10} (msec) Ni^{61}	ν (Mc/sec) Fe^{57}	ν (Mc/sec) Ni^{61}	$\nu^2 T_{10}$ (Mc^2/sec) Fe^{57}	$\nu^2 T_{10}$ (Mc/sec) Ni^{61}
300	0.9	0.7	45	85	1.8	5.1
77	0.7	1.7	47	89	1.5	13.5
4	10.0	6.0	47	89	22.1	47.5

T_{10} for Ni^{61} in pure nickel and 0.5% Co^{59} in 99.5% nickel.

Temp (°K)	T_{10} (msec)[a] Ni^{61}	T_{10} (msec) Co^{59}	ν (Mc/sec) Ni^{61}	ν (Mc/sec) Co^{59}	$\nu^2 T_{10}$ (Mc^2/sec) Ni^{61}	$\nu^2 T_{10}$ (Mc^2/sec) Co^{59}
300	0.35	0.05	26	112	0.24	0.63
77	1.5	0.13	28	120	1.2	1.9
4	15	1.4	28	121	11.8	20.5

[a] See Ref. 2.

[b] The resonance frequencies are rounded to the nearest megacycles. See A. C. Gossard, Ph.D. thesis, University of California, 1960 (unpublished); J. I. Budnick, L. J. Bruner, R. Blume, and E. L. Boyd, J. Appl. Phys. **32**, 120S (1961) and Refs. 4–7.

alloy tend to be decreased compared with those in the pure solvent metal.

Although in Table II the T_{10} for the solute nuclei is compared with the T_{10} in the pure host nickel, it would be better to actually compare the relaxation times for the nuclei of the solute and solvent atoms in the same sample. In the case of 0.5% cobalt in nickel at 77°K we have done this. As already discussed (see Sec. IV.A), T_{10} for the Ni^{61} in a sample annealed for one hour at 1100°C was 1.3 msec, while for Co^{59} the value of T_{10} in the same sample was 0.13 msec. From Eq. (4) we see then that $\langle\theta^2\rangle\tau_c$ is about one-half as large for the Co^{59} as for the Ni^{61}, and this probably represents a somewhat smaller angular displacement of the cobalt spin in the domain wall.

In the 0.5% cobalt in nickel sample, the Co^{59} relaxation times in domain walls were found to be nearly independent of annealing time. This seems to indicate that in this sample the solute atoms themselves rather than the degree of anneal are limiting the wall motion.

D. Discussion of T_1 in the Domains

The longer times T_{1M} given in Table I are nearly inversely proportional to temperature and are believed to approach the limiting value of T_1 appropriate to the nuclei in domains and to be determined by relaxation through the conduction electrons. The quantity $1/\gamma_n^2 T_{1M}T$ is a measure of the strength of the conduction electron relaxation mechanism normalized for the difference in nuclear gyromagnetic ratios. In Table III we compare $\gamma_n^2 T_{1M}T$ for the nuclei on the impurity atoms with the corresponding quantity for the nuclei of the pure host metal using the value of T_1T measured in the pure metals by Weger.[3] We also list in the same table the atomic magnetic moments measured by neutron diffraction.[13,14]

The quantity $\gamma_n^2 T_{1M}T$ is seen in all cases to be greater for the impurity nucleus than for the nucleus of the corresponding pure host metal and this is particularly true for the case of 0.5% Co^{59} in nickel. Also, especially in the case of Ni^{61} in iron where the signal at high power levels was weak and the dependence of T_1 on the power level was quite strong, we cannot be sure we have measured the longest time so that the T_{1M} is only a lower limit for the value of T_1 in the domains.

The longer relaxation times for the impurity nuclei can probably be understood qualitatively as representing a partial decoupling of the spin on the impurity

TABLE III. A comparison of the quantity $T_{1M}T$ for the impurity nuclei in the dilute alloys (from Table I) with the corresponding quantity in the pure host metal, as measured by Weger.[a] The quantities $\gamma_n^2 T_{1M}T$ are also compared.[b] The atomic moments[c] in Bohr magnetons (bm) are also given.

Nucleus	μ (bm)	$T_{1M}T$ (sec°K)	γ_n (cps G^{-1})	$\gamma_n^2 T_{1M}T$
Co^{59} in Co	1.7	0.08	6.27×10^3	3.1×10^6
1% Ni^{61} in 99% Co	0.6	1.0	2.38×10^3	5.7×10^6
Fe^{57} in Fe	2.2	2.5	0.864×10^3	1.9×10^6
1% Ni^{61} in 99% Fe	1.0	1.2	2.38×10^3	6.8×10^6
Ni^{61} in Ni	0.6	0.115	2.38×10^3	0.65×10^6
0.5% Co^{59} in 99.5% Ni	1.7	0.3	6.27×10^3	11.8×10^6

[a] See Ref. 3.

[b] For values of γ_n see Freeman *et al.*, Proc. Roy. Soc. (London) **242**, 455 (1957) for Co^{59}; P. R. Locher and S. Geschwind, Phys. Rev. Letters **11**, 333 (1963) for Ni^{61}; and C. W. Ludwig and H. H. Woodbury, Phys. Rev. **117**, 1286 (1960) for Fe^{57}.

[c] See Refs. 13 and 14.

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[13] M. F. Collins and J. B. Forsyth, Phil. Mag. 8, 401 (1963).

[14] M. F. Collins and D. A. Wheeler, Proc. Phys. Soc. (London) 82, 633 (1963).

atom from the spin-wave spectrum; that is, a spin flip of the impurity atom spin, which is different from that of the host (see Table III), will probably in general require a higher energy than for the case of the pure metal. This probably reduces the enhancement of the conduction electron relaxation by the spin waves.

Note also that while $\gamma^2 T_{1M} T$ for Ni^{61} in cobalt and iron is longer than in pure nickel, it is still shorter than $\gamma^2 T_1 T$ for the case of a nonferromagnetic metal like Cu^{63} in copper where $\gamma^2 T_1 T$ is about 45×10^6 (with γ in $cpsG^{-1}$ and $T_1 T$ in sec °K).[15] Similarly, while $\gamma^2 T_{1M} T$ for Co^{59} in nickel is longer than for Co^{59} in pure cobalt, it is still less than $\gamma^2 T_1 T$ for V^{51} in vanadium, where $\gamma^2 T_1 T$ is about 39×10^6.[16]

E. Discussion of T_2

In the case of Ni^{61} in cobalt, T_2 is being determined by nuclear spin-spin interactions. We will discuss various coupling mechanisms. The Suhl-Nakamura interaction[17,18] involving virtual excitation of spin waves is a nuclear spin-spin coupling mechanism for Co^{59} in pure cobalt.[1] This mechanism, however, at least in the pure ferromagnets and at low temperatures (well below the Curie point), couples nuclear spins according to $I_1^+ I_2^- + I_2^+ I_1^-$ and this would not be a broadening mechanism for unlike nuclear spins which would not be precessing at the same Larmour frequency.[18] This would probably to a large extent be true also for the case of 1% Ni^{61} in cobalt, where the spin-wave excitations are still mainly transverse.

The Ruderman-Kittel[19] interaction involving a scalar coupling between nuclear spins would be a broadening mechanism for unlike spins. One might argue that the same mechanism which leads to the short values of $T_1 T$ for both Co^{59} and Ni^{61} in the 1% Ni^{61} in cobalt alloy would also lead to an enhanced Ruderman-Kittel interaction. However, an enhanced interaction of this sort involving spin-wave excitations would at low temperatures be anisotropic in the same way as the Suhl interaction and would not be effective in broadening unlike spins. However, the ordinary Ruderman-Kittel interaction is about the right value to explain the observed T_2. This can be seen as follows: In the case of Cu^{63} in copper metal, the experimental results could be explained by taking the scalar indirect exchange coupling A/h between two nearest Cu^{63} neighbors to be about 420 cycles or three times the calculated value.[15] Assuming a similar coupling between the nickel and cobalt nuclei leads to an exchange coupling between neighbors of

$$420\gamma(Co^{59})\gamma(Ni^{61})/[\gamma(Cu^{63})]^2=120 \text{ cycles.}$$

The nearest-neighbor contribution to the Van Vleck second moment is then given by

$$\langle\Delta\nu^2\rangle=4I(I+1)(A/h)^2.$$

This gives a width $[\langle\Delta\nu^2\rangle]^{1/2}$ of about 1 kc/sec or T_2 about 0.2 msec. Although the above is the order of magnitude of the T_2 observed, the fast spin exchange of the Co^{59} nuclei would tend to narrow the line and make the shape more nearly Lorentzian so that an exact calculation of T_2 is difficult.

In the case of Ni^{61} in iron and Co^{59} in nickel (where we believe we can neglect nuclear spin-spin interaction) the T_2 relaxation mechanism is evidently related to T_1 at high temperatures in the domain walls. At low temperatures, particularly in the domains, T_2 may be due to the mechanism discussed by Moriya. However, the situation is not completely understood.

V. CONCLUSION

The longitudinal relaxation times for the nuclei at the solute atoms in the domain walls in these alloys can be understood as due to the motion of the domain walls. However, it appears that the relaxation time for the solute nuclei will generally be somewhat longer than the relaxation time for the nucleus at the host atom due to the pinning of the domain wall near the solute atom. In the domains it appears that the coupling of the nucleus to the conduction electron which determines T_1 at high power levels is weaker for the solute atom than for the host atom in these materials. In the case of Ni^{61} in cobalt, T_2 is being determined by nuclear spin-spin interaction. A Ruderman-Kittel indirect coupling of nuclei by conduction electrons seems to account for the observed value of T_2.

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