NMR enhancement of a modulating field due to the anisotropic component of the hyperfine field in hcp Co and YCo_5

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Modulation of the ^{59}Co NMR spin-echo envelope in hcp Co and YCo_5 results from the application of a small 80-kHz field. The modulating field is enhanced parallel to the local axis of quantization by an enhancement factor $\eta_{\parallel}$ which varies with position in the domain wall. The maximum enhancement factor is found to be ~ 80 in hcp Co and ~ 1000 in YCo_5. When $\eta_{\parallel}$ is taken to arise from an anisotropic component of the hyperfine field it is estimated that this anisotropic component has a magnitude of 5.7 kOe in hexagonal Co and 60 kOe in YCo_5.

I. INTRODUCTION

Several workers[1-3] have reported the observation of two NMR peaks arising from a single site in magnetically ordered material containing domain walls. It has been suggested that one peak comes from nuclei located at domain-wall edges while the other comes from nuclei at the center.[3-5] The origin of this effect is an anisotropic contribution H_a to the hyperfine field, which leads to a resonance frequency which varies with the direction of the electronic moment and hence with the position of the nucleus in the wall. Either an anisotropic electronic spin density in the outer electronic shell, or an anisotropic g factor (orbital contribution) can lead to this effect in Co (see Ref. 3); in YCo_5 the origin of H_a could be more complicated. We have observed a modulation of the NMR spin-echo amplitude from ^{59}Co in hexagonal Co and YCo_5, by the application of an ac modulating field, which strongly supports this suggestion. This follows because the modulating field is enhanced by the anisotropic contribution to the hyperfine field as the domain-wall motion shifts the effective position of the nuclei.

II. EXPERIMENTAL RESULTS

The samples consisted of 50-μm-diam particles of Co and 10–100-μm-diam particles of YCo_5, both magnetically aligned parallel to the c axis in a wax binder. The experimental data were taken at 77 K for Co and at 1.5 K for YCo_5. The NMR spin echo, following a two-pulse sequence, was observed with and without a modulating field, $\vec{H}_m$, applied parallel to the pulsed rf field $\vec{H}_1$.

The NMR signal originates entirely from domain walls since no echo was observed with $\vec{H}_1$ perpendicular to the c axis, while a relatively strong echo was observed with $\vec{H}_1$ parallel to the c axis. This is consistent with the strong uniaxial magnetocrystalline anisotropy in these materials,[6,7] which leads to a very small enhancement factor within the domains.

Figure 1 shows the relative spin-echo intensity for hexagonal cobalt, extrapolated to $\tau = 0$, where τ is the pulse separation, as a function of frequency. The spectrum shows a double-peaked structure similar to that reported by other investigators.[3,8]

The echo-decay envelope appears in Fig. 2(a), along with the echo-decay envelope with a modulating field of amplitude $H_m = 1.4$ Oe and frequency $\nu_m = 80$ kHz applied along the c axis. The corresponding echo decay envelopes for YCo_5 are shown in Fig. 2(b). It is apparent that the echo amplitude

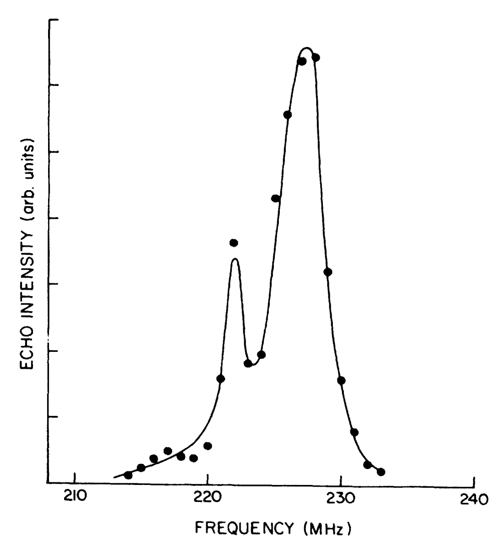

FIG. 1. Spin-echo intensity as a function of frequency for hexagonal cobalt at 77 K.

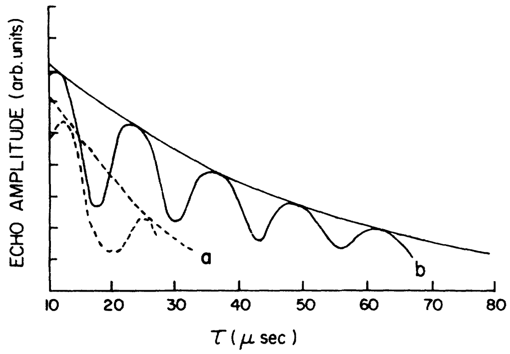

FIG. 2. Echo-decay envelopes with and without an 80 kHz modulating field, H_m, applied along the c axis; (a) hcp cobalt at 224 MHz; $H_m = 1.4$ Oe (b) YCo_5 at 124 MHz; $H_m = 0.1$ Oe.

is modulated with a period equal to $1/\nu_m$ and has maxima at $\tau = n/\nu_m$, where τ is the pulse separation and n is an integer.

The modulation was strongest in the center of the spectrum and weakest at the high- and low-frequency edges, for a constant H_m. Figure 3 is a plot of $A_{min}(H_m)/A_0$ as a function of frequency, for hexagonal Co where $A_{min}(H_m)$ is a minimum of the modulated echo intensity and A_0 is the unmodulated echo intensity measured at the same pulse separation τ; H_m and ν_m were maintained at 1.4 Oe and 80 kHz through the entire frequency range. The striking feature shown by these data is that $A_{min}(H_m)/A_0 \approx 0.9$ at the low-frequency end of the spectrum, drops through a minimum of ~0.1 near the center and again rises monotonically to large values near the high-frequency edge.

Figure 4 shows the relative spin-echo intensity for YCo_5, again extrapolated to $\tau = 0$, as a function of frequency without a modulating field. This result is similar to that reported by Streever[9] for

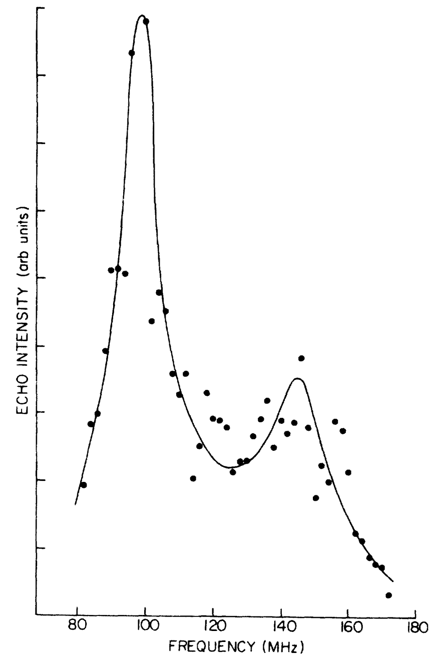

FIG. 4. Spin-echo intensity as a function of frequency for YCo_5 at 1.5 K. Because of the broad spectrum, the experimental echo amplitudes at each frequency ν have been divided by ν^2 to correct for the fact that the induced nuclear signal and the nuclear polarization are both proportional to ν.

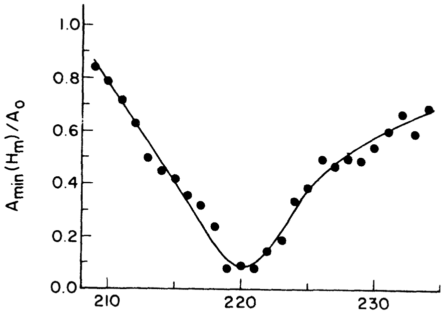

FIG. 3. $A_{min}(H_m)/A_0$ as a function of frequency for hexagonal cobalt, where $A_{min}(H_m)$ is a minimum of the modulated echo intensity and A_0 is the unmodulated echo intensity measured at the same pulse separation.

$NdCo_5$; however, it should be stressed that at low temperatures the spins in the domains are in the c plane for $NdCo_5$, and along the c axis for YCo_5 and one would expect a quantitative difference between the two spectra. The width of the spectrum in YCo_5 is 60–70 MHz which we will assume is predominantly due to broadening by an anisotropic contribution to the hyperfine field.

Figure 5 is the corresponding plot of $A_{min}(H_m)/A_0$ as a function of frequency. H_m and ν_m were here maintained at 0.1 Oe and 80 kHz throughout the entire NMR frequency range. The plot of $A_{min}(H_m)/A_0$ against frequency shows the same basic shape as was obtained for hexagonal cobalt. However, there does appear to be some additional structure which could result from the two nonequivalent Co sites in YCo_5. At its minimum $A_{min}(H_m)/A_0 \approx 0.2$.

III. INTERPRETATION

The experimental data shown in Fig. 2(b) are very similar to the direct electron-spin-echo modulation reported by Dupont and Woonton,[10] and Srivastava[11] who showed that the effect was related to the ratio of the magnitude of the modulating field h to that of the rf field during the pulse H_1. According to their calculations,

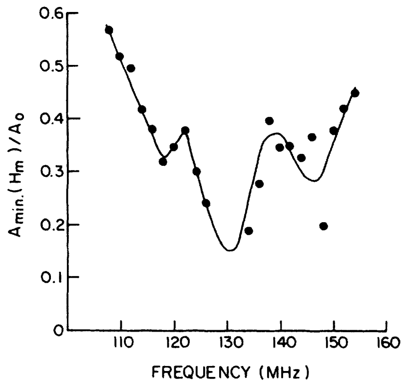

FIG. 5. $A_{min}(H_m)/A_0$ as a function of frequency for YCo_5.

$$A_{min}(H_m)/A_0 \sim 0.1 \quad \text{when } h/H_1 \to 1.2 . \qquad (1)$$

We will estimate H_1 for a 180° pulse, assuming a 90°–180° pulse sequence. In a rotating coordinate system one may then write $\pi = \gamma_N H_1 t_1$, where γ_N is the gyromagnetic ratio for ^{59}Co, and t_1 is the pulse width. Using $\gamma_N/2\pi = 1.01 \times 10^3$ Hz/Oe and $t_1 = 5\mu$ sec one obtains

$$H_1 \simeq 100 \text{ Oe} \qquad (2)$$

for a 180° pulse. Clearly, H_m is not directly modulating the spin-echo envelope since $H_m/H_1 = 1.4/100$, or two orders of magnitude too small to make any direct effect of H_m experimentally detectable. We therefore require an enhancement factor $\eta \approx 80$ between the external modulating field and the effective field seen at the nuclei in the domain walls. We will assume that this arises out of an anisotropic contribution to the hyperfine field varying through the domain wall, as in hexagonal Co, of the form[3]

$$H_a(x) = H_a \sin^2(\pi x/\delta) , \qquad (3)$$

where δ is the wall width and x is a position in the wall referred to one "edge". Then the enhancement factor for the modulating field parallel to a local quantization direction in the domain wall may be written as

$$\eta_{\parallel} = \frac{h}{H_m} = \frac{\Delta H_a}{H_m} \simeq \frac{\partial H_a}{\partial x}\frac{\Delta x}{H_m}, \qquad (4)$$

where h is the effective modulating field at the nucleus, and Δx is the change in wall location when H_m goes through a half period to satisfy the condition for a minimum in the echo amplitude. One may also write the change in magnetization as

$$m_{\parallel} = \chi_{\parallel} H_m = M_0 \frac{2\Delta x}{W}, \qquad (5)$$

where $\chi_{\parallel}$ is the magnetic susceptibility parallel to the c axis (almost entirely due to domain wall motion at 77 K), W is the average spacing between walls, and M_0 is the saturation magnetization. Using Eqs. (3)–(5) we obtain

$$\eta_{\parallel} = (\pi W \chi_{\parallel} H_a / 2\delta M_0) \sin(2\pi x/\delta) . \qquad (6)$$

The maximum $\eta_{\parallel} = \eta_{max}$ occurs when $x = \frac{1}{4}\delta$, giving

$$\eta_{max} = \pi W \chi_{\parallel} H_a / 2\delta M_0 . \qquad (7)$$

The maximum enhancement factor is experimentally located at $\nu_0 = 220$ MHz as seen in Fig. 3. Since the observed depth of modulation of the echo envelope suggest $\eta_{max} \simeq 80$, we can solve Eq. (7) for H_a, the required anisotropy in the hyperfine field. Using the values for the saturation magnetization $M_0 = 1420$ emu/cm^3 and the domain-wall energy $\gamma = 10.7$ erg/cm^2 for cobalt (taken from Ref. 12), we estimate values for the domain width and wall thickness in 50-μm particles as described in Ref. 13. Here we have assumed that the result which is given for a plate of thickness T will not be much different from that of a particle of the same diameter. We obtain $W = 1.25$ μm and $\delta = 350$ Å. The low-field ac susceptibility measured at 77 K and 15 kHz is $\chi_{\parallel} = 0.32$ emu/cm^{3}Oe. We then calculate $H_a = 5.7$ kOe. For comparison, we see in Ref. 3 that in hexagonal cobalt the dipolar component of the anisotropic hyperfine field is 1.5 kOe and the orbital component is 5.4 kOe resulting in a total anisotropy of approximately 6.9 kOe. Using our value of H_a we expect to observe a line width

$$\Delta\nu \simeq (\gamma_N/2\pi) H_a = 1.01 \times 10^3 \times 5.7 \times 10^3 = 5.8 \text{ MHz} .$$

The observed linewidth for hexagonal cobalt in Fig. 1 is $\delta\nu \simeq 8$ MHz.

The maximum enhancement factor in YCo_5 is similarly estimated to be $\eta_{max} \simeq 1000$ at $\nu_0 = 130$ MHz in Fig. 5. If we assume that the form of the anisotropic contribution to the hyperfine field through the domain wall is similar to that in cobalt we can again calculate H_a. We take $W = 1.7$ μm and $\delta = 55$ Å from Ref. 13, and $M_0 = 901$ emu/cm^3 from Ref. 14; the low-field ac susceptibility measured at 4.2 K and 15 kHz is $\chi_{\parallel} = 1.6 \times 10^{-2}$ emu/cm^{3}Oe. Then we have $H_a = 57$ kOe, and thus we should expect to observe an anisotropically broadened linewidth of 58 MHz. This compares favorably with the observed linewidth of $\Delta\nu \simeq 60$–70 MHz.

IV. CONCLUSIONS

Considering the good agreement between the calculated and observed linewidths, the origin of the observed modulation can convincingly be traced to an anisotropy in the hyperfine field, which causes a broadening of the NMR spectrum across the domain walls. This is true not only for hexagonal Co which contains only one site, but also for YCo_5 which contains two nonequivalent ^{59}Co sites. This result implies that the two site resonances almost completely overlap and that the two NMR peaks observed in YCo_5 come from nuclei located at the domain wall edges and at the domain wall center. Our conclusion is consistent with the fact that some additional structure in the modulation versus frequency curve was observed for YCo_5 which it is believed is due to the presence of the two slightly different Co sites.

The modulation of the spin-echo decay envelope via the longitudinal enhancement factor $\eta_{\parallel}$ should be observed in any domain-wall-driven NMR spin-echo experiment. This effect does require a large anisotropic contribution to the hyperfine field combined with a relatively narrow domain wall. Kubo *et al.*[15] have shown, using a single crystal of manganese ferrite, that Mn^{3+} located at the B site has a large anisotropic component in its hyperfine field. We have confirmed the presence of an enhanced spin-echo modulation for Mn^{3+} in manganese ferrite at 1.5 K.

The spin echo could also be very simply modulated if the modulating field were large so that the movement of the domain wall, in the time between rf pulses, would be sufficient to remove a significant fraction of the nuclear spins, excited by the first pulse, from the domain wall. A simple calculation rules out this effect since the ratio of the domain-wall motion between pulses to domain-wall width is $\sim 10^{-4}$, for the modulating fields used in the present experiment.

The ordinary domain-wall enhancement factor η also depends on the position within the wall.[16] This could also lead to an echo envelope modulation. However, the effective change in η between pulses, $\delta\eta$, would be on the order of the total change $\Delta\eta$ through the domain wall, times the ratio of domain wall displacement between pulses to domain wall width. With $\Delta\eta \sim 10\eta$[16] we have $\delta\eta = 10\eta \times 10^{-4} = \eta \times 10^{-3}$ and this rules out this possibility.

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[1]H. Nagai, T. Hihara, and E. Hirahara, J. Phys. Soc. Jpn. 29, 622 (1970).

[2]E. Koster and B. G. Turrell, Phys. Lett. A 39, 211 (1972).

[3]M. Kawakami, T. Hihara, Y. Koi, and T. Wakiyama, J. Phys. Soc. Jpn. 33, 1591 (1972).

[4]M. A. Butler, Phys. Rev. B 8, 5122 (1973).

[5]M. A. Butler, Int. J. Magn. 4, 131 (1973).

[6]G. Hoffer and K. Strnat, IEEE Trans. Magn. 2, 487 (1966).

[7]W. Sucksmith and J. E. Thompson, Proc. R. Soc. A 225, 362 (1954).

[8]S. G. Bailey, D. C. Creagh, and G. V. H. Wilson, Phys. Lett. A 44, 229 (1973).

[9]R. L. Streever, AIP Conf. Proc. 24, 462 (1975).

[10]M. Dupont and G. A. Woonton, Can. J. Phys. 46, 87 (1968).

[11]V. C. Srivastava, J. Appl. Phys. 39, 5837 (1968).

[12]J. Kaczer, Sov. Phys.-JETP 19, 1204 (1964).

[13]J. D. Livingston and M. D. McConnell, J. Appl. Phys. 43, 4756 (1972). (These are representative values obtained at room temperature and were used for our order-of-magnitude estimation of H_a at 4.2 K).

[14]W. G. D. Frederick, Ph.D. dissertation (University of Cincinnatti, 1973) (unpublished).

[15]T. Kubo, A. Hirai, and H. Abe, J. Phys. Soc. Jpn. 26, 1094 (1969).

[16]C. H. Cobb, V. Jaccarino, M. A. Butler, J. P. Remeika, and H. Yasouka, Phys. Rev. B 7, 307 (1973).