ISSN 0029 - 3865

CBPF-NF-052/86

NMR STUDY OF THE CHANGE IN THE DIRECTION OF MAGNETIZATION OF $HoCo_2$†

by

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*Supported by CNPq (Brasil)

†to be published in J. Appl. Phys.

ABSTRACT

The intermetallic compound $HoCo_2$ is magnetically ordered and changes its direction of magnetization around 15 K, as the temperature is increased, from [110] to [100].

We report a ^{59}Co pulsed NMR study of $HoCo_2$ in the temperature range 4 K - 65 K. The NMR spectra show two peaks at low temperature and one peak at high temperatures, as expected from a model whereby the magnetic equivalence of the cobalt sites is determined by the direction of magnetization. This change in the spectra can be simulated from lattice sums of the magnetic dipolar fields acting on the cobalt sites.

The present results are consistent with a change of direction of magnetization from [110] to [100] through intermediate directions contained in the plane z=0.

Key-words: NMR; $HoCo_2$

1. Introduction

The intermetallic compound $HoCo_2$ cristallizes in the cubic Laves structure (C15). It is magnetic with an ordering temperature of 77 K[1]. Magnetization studies[2] of $HoCo_2$ single crystals show a change in the direction of magnetization around 15 K. This is also observed with specific heat measurements[3] and resistivity studies[4] in polycrystalline samples. The spin re-orientation process can be described within a simple model where the Hamiltonian for the system contains a magnetic interaction with the molecular field and a crystal-field interaction[4].

2. Experimental

The $HoCo_2$ ingots were prepared in an rf furnace with a 6% excess of holmium, and subsequently annealed at 900 C for 72 h; the X-ray analysis detected no foreign phases. For the present experiment, the ingots were ground to a powder which was then mixed with silicone oil. The NMR measurements were made using a Bruker SXP spectrometer and a helium flow cryostat.

The experimental spectra were drawn by plotting spin echo height versus frequency (Fig. 1). Pulses of 0.5 microseconds were used, with a separation of 10-15 microseconds and a repetition rate of under one kHz.

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3. Results

The NMR spectra (Fig. 1) show at 4.2 K two lines of roughly the same intensity (area). The separation between the lines decreases with increasing temperature, and above 20 K only one line is visible. The spectra could not be measured above 65 K since the echoes become then too small and the signal to noise ratio very unfavourable. In Fig. 2 the positions of the lines of the spectra are plotted as a function of temperature.

The spectrum of $HoCo_2$ at 4.2 K obtained in the present study, consisting of two lines with approximately the same area (Fig. 1), differs from the results obtained by Hirosawa and Nakamura[5] and Rubinstein et al[6]. These authors show more complex spectra, where two lines appear superposed on a broad background. The structure observed by Hirosawa and Nakamura disappears with an external applied field, and was therefore attributed to nuclei in domain walls. The excitation conditions used in our experiments must have favoured the observation of ^{59}Co echoes from the domains.

4. Discussion

The correlation between direction of magnetization and number of peaks in the spectrum is in principle

easy to be established: the four cobalt atoms that form tetrahedra in the C15 structure are equivalent from the crystallographic point of view, but the magnetic dipolar fields due to the atomic moments on each site may be different, depending on the direction of magnetization. Thus, in the case of magnetization parallel to a [110] direction the dipolar fields are equal for each pair of atoms, and therefore the spectrum shows only two lines. In the case of [100] all four fields are identical, and only one line appears, and in the [111] case there are two lines with intensities in the ratio 3:1[7].

We might obtain a spectrum with three lines if the magnetization is contained, for example, in the plane x=y, along a direction intermediate between the directions [111] and [110]. Using the dipole lattice sums for the C15 structure[7], we compute the dipolar fields at the four sites, for any direction of magnetization. In the case of a magnetization that turns from [110] to [001], remaining in the plane x=y, the positions of the lines are such that the spectrum changes from two lines to three lines, two lines again, and then to a single line.

A spectrum with two equal lines, such as found in the present work at 4.2 K (Fig. 1) is obtained when the magnetization is parallel to a [110] direction, or more generally, to any direction in the plane z=0, excluding the

[100] direction (Fig. 3). The magnetization studies, however, confirm that the direction is indeed [110] at low temperature[2].

The series of results presented here (Fig. 1) confirm that the direction of magnetization in $HoCo_2$ changes as a function of temperature. The spectra show two widely separated (by 19 MHz) lines below 18 K, two partly resolved lines at 20 K and one single line above that temperature (Fig. 1). The line positions versus temperature are shown in Fig. 2; they reflect a change in the direction of magnetization with increasing temperature, from [110] to [100], but the change is not as sharp as previously observed with other techniques[1-3].

One can infer from Fig. 2 the trajectory followed by the magnetization as it changes from a [110] direction to [100]. As mentioned above, some trajectories could lead to spectra with three peaks, not observed experimentally. From the experimental curve of peak position versus temperature (Fig. 2) one then concludes that the path of the magnetization is contained in the plane z=0. The position of the peaks computed from the lattice sums using $\mu_{Co}=1\ \mu_B$ and $\mu_{Ho}=10\ \mu_B$ are given in Fig. 3. Based on the separation between peaks as a function of the angle and assuming $\phi=45$ degrees for the spectrum at 4.2 K (direction [110]), we estimate that at 20 K the magnetization is still about 3 de-

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grees from the [100] direction, from the ratio of peak separation at these two temperatures.

The ^{59}Co NMR results obtained in the present work are qualitatively similar to those obtained[8] using Mössbauer Spectroscopy on ^{57}Fe diluted in $HoCo_2$; the magnitude of the splittings is however, different.

Figures Captions

Fig. 1 ^{59}Co NMR spectra of $HoCo_2$ versus frequency, for different temperatures. The change in the spectra from two lines to one single line reflects the change in the direction of magnetization, from [110] to [100].

Fig. 2 Resonance frequencies (peak positions) of the ^{59}Co NMR of $HoCo_2$, as a function of temperature. The lines were drawn to guide the eyes.

Fig. 3 Computed resonance frequencies of ^{59}Co in $HoCo_2$ for directions of magnetization contained in the plane z=0, as a function of angle ϕ. The lines were calculated from magnetic dipolar sums at the cobalt sites.

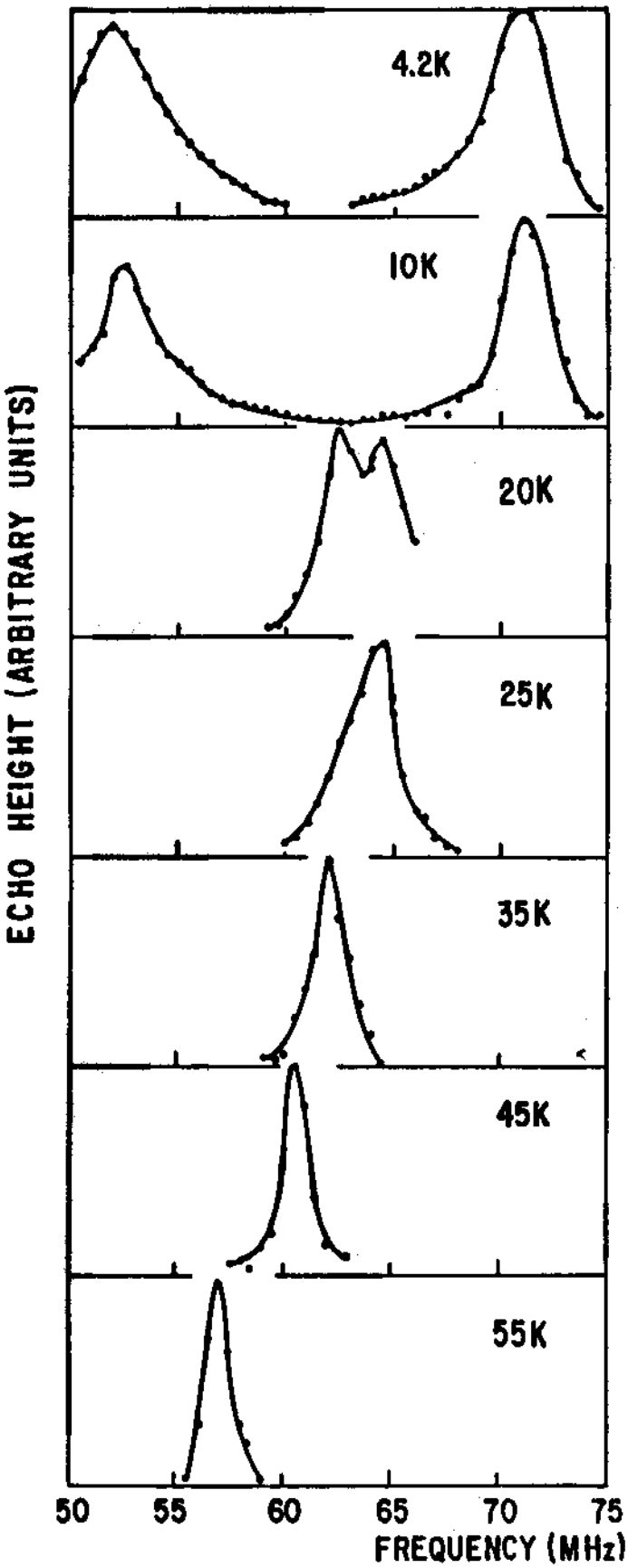
4.2K
10K
20K
25K
35K
45K
55K
ECHO HEIGHT (ARBITRARY UNITS)
50
55
60
65
70
75
FREQUENCY (MHz)

Fig. 1

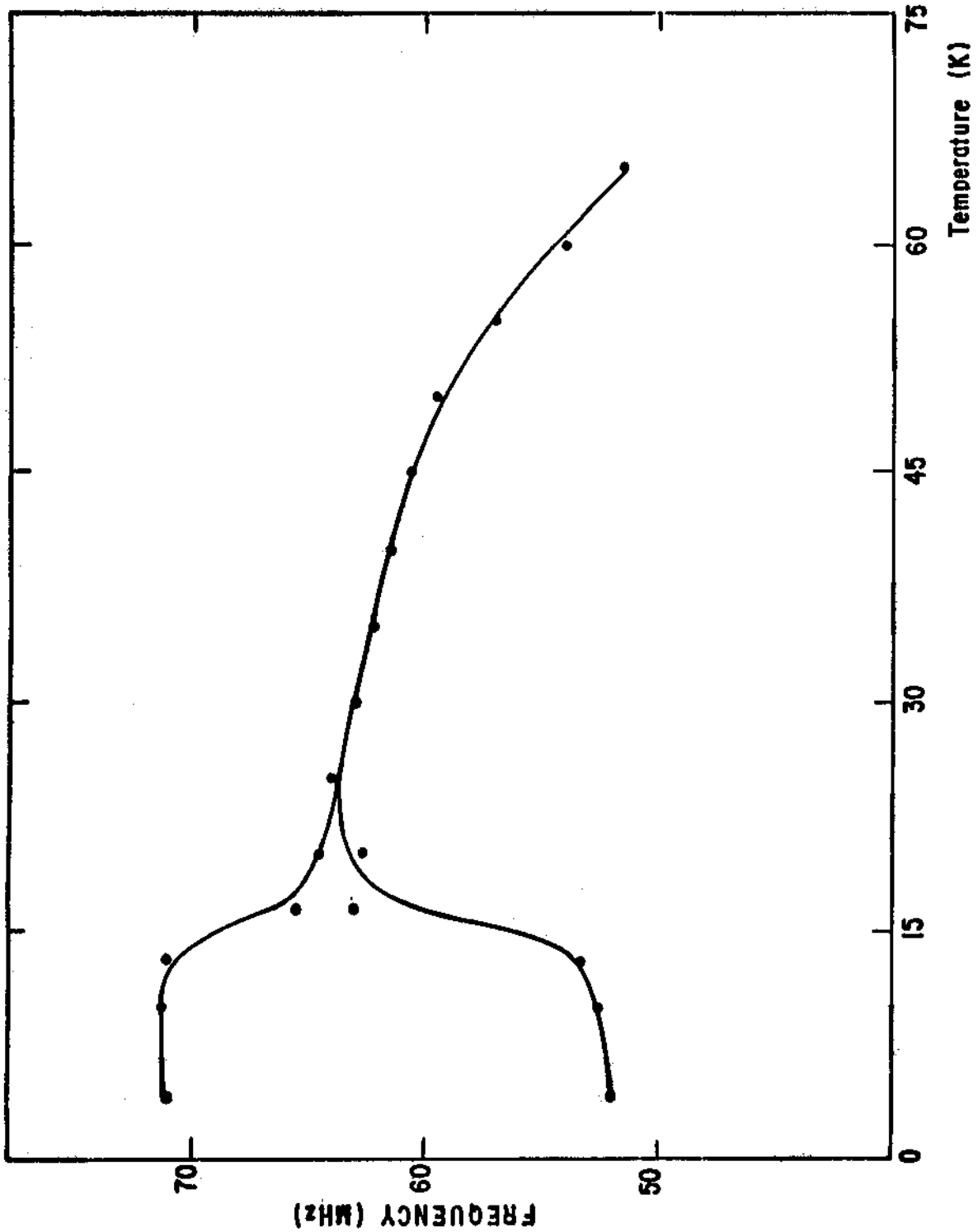

Fig. 2

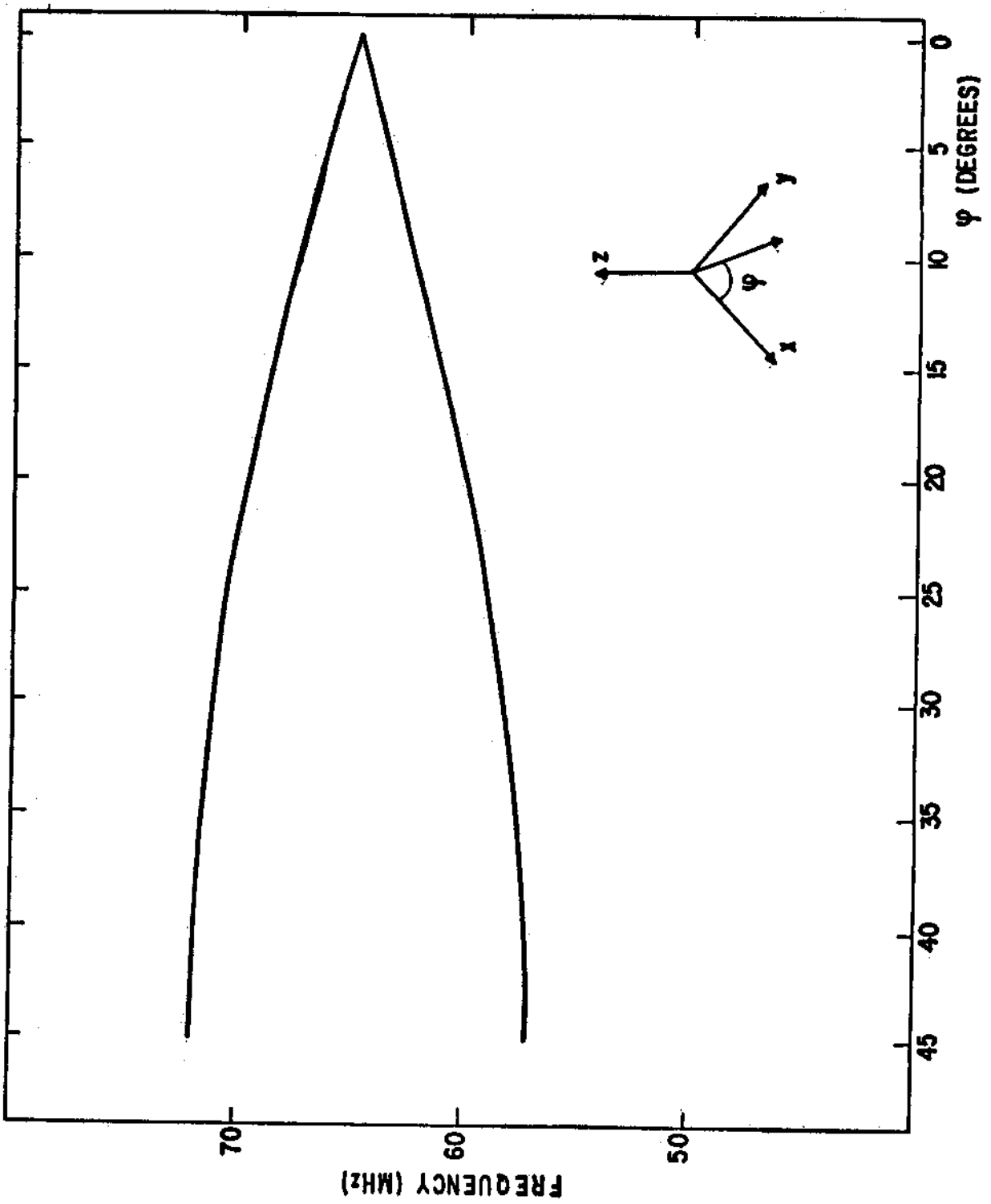

Fig. 3

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