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Publication Date
1970-02-01

Submitted to Physics Letters

UCRL-19561
Preprint

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AEC Contract No. W-7405-eng-48

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PERTURBED ANGULAR CORRELATIONS IN FERROMAGNETS: THE ^{100}RhNi CASE*

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Studies of the hyperfine field for the system ^{100}RhNi by both NMR/PAC and TDPAC techniques are reported and compared. The value at 296.0(5) K is $|H_{hf}| = 207.1(6)$ kG. An upper limit on inhomogeneous and relaxation broadenings is set at 2.5(5) MHz.

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When an intermediate state in an angular correlation cascade interacts with a magnetic field by dipole coupling,

$$\mathcal{H} = -\vec{\mu}\cdot\vec{H} = -\gamma\vec{I}\cdot\vec{H} \quad , \tag{1}$$

the nuclear moment precesses about $\vec{H}$ at the Larmor frequency

$$2\pi\nu_L = \gamma H \quad , \tag{2}$$

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*This work was performed under the auspices of the U. S. Atomic Energy Commission.

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where γ is the nuclear gyromagnetic ratio. Consequently the angular correlation pattern, which for dipole transitions has the form

$$W(\theta) = 1 + A_2 P_2 (\cos \theta) \quad , \tag{3}$$

becomes

$$W(\theta,t) = 1 + A_2 P_2 (\cos [\theta - 2\pi\nu_L t]) \tag{4}$$

for the magnetic field $\vec{H}$ perpendicular to the detector plane, and

$$W(\theta,t) = 1 + (1/5)\, A_2 (1 + 2 \cos 2\pi\nu_L t + 2 \cos 4\pi\nu_L t)\, P_2 (\cos \theta) \tag{5}$$

for a randomly oriented magnetic field [1] of constant magnitude H.

The accurate determination of ν_L is a problem of central importance in the application of perturbed angular correlations to solid state physics. In this letter we describe two ways in which the techniques for determining ν_L with high accuracy for the test case ^{100}RhNi have been improved recently.

A. Nuclear Magnetic Resonance/Perturbed Angular Correlations (NMR/PAC)

This technique was first demonstrated in this Laboratory in 1966 on the ^{100}RhNi case [2]. While qualitatively successful, the experiment left several questions unanswered: 1) the observed 2% effect was smaller than expected and 2) the resonance linewidth of 20 MHz (fig. 1(a)) was far greater than the 0.94 MHz natural linewidth dictated by the intermediate state mean life, τ_N = 339 ns. In further studies [3], the asymmetric line was resolved into two components (fig. 1(b)).

We have now established that both effects were mainly attributable to inhomogeneous broadening caused by Pd impurity in the Rh metal used as target

material to produce ^{100}Pd via the ^{103}Rh(p,4n)^{100}Pd reaction. This Pd impurity went through chemical separation with the ^{100}Pd activity and appeared as a 1% impurity in the Ni samples. From Budnick's work [4] we would expect that, for a ^{100}Pd atom within 2 or 3 lattice positions of a Pd impurity atom in a nickel lattice, a resonance at a frequency ≈5% below the main line would appear. Substantial line broadening would also be expected. The early spectra are fully consistent with Budnick's results for an alloy with 1% impurity.

In a new series of experiments, higher purity alloys were prepared and the temperature and rf field intensity were more carefully controlled. The resonance lines were much narrower and the NMR/PAC effect was greater. The results are shown in fig. 1(c). Resonance occurs at 338 MHz, the narrowest linewidth observed being 4 MHz.

We conclude that in ferromagnets NMR/PAC can produce resonance lines which are essentially equivalent to those observed by conventional NMR.

B. Time-Differential Perturbed Angular Correlations (TDPAC)

Because $W(\theta)$ has period π, the modulation frequency of $W(\theta,t)$ in eq. (3) is $2\nu_L$. In the case of eq. (4) the modulation pattern is more complex [1], containing both ν_L and $2\nu_L$. For the ^{100}RhNi system at room temperature, ν_L = 340 MHz, so that with an external field the coincidence rate oscillates through a full cycle in only 1.5 ns, whereas a component with period $1/\nu_L$ = 3 ns is also present in an unpolarized source. Direct observation of these high frequencies, particularly for the low energy cascade 84.0 keV - 74.8 keV, requires much better detector timing characteristics than have been available with the usual Ge(Li) and NaI(Tl) systems.

The best figure we obtained by using Ge(Li) detectors was 6.3 ns FWHM for prompt coincidences at 84 keV - 75 keV. This, of course, is too slow for our case. An apparatus was built employing Sn-loaded plastic scintillators NE140, together with RCA 8850 photomultiplier tubes. The Sn content in the phosphor enhances the full energy peak detection at lower energies and this peak, together with the Sn escape peaks, is clearly visible. The (unresolved) full energy peaks associated with the 84-keV - 75-keV cascade could be observed directly and a fast resolving time of 1.20 ns FWHM for prompt coincidences at these energies was achieved. With this system, we were able to resolve even the $2\nu_L$ = 680 MHz modulation component, producing hundreds of oscillations over the useful range of about three mean lives of the 75-keV level. Of course, the amplitude of these oscillations was substantially reduced by the finite resolving time of the coincidence circuit.

The following analysis refers only to unpolarized experiments. A part of a time-differential curve with detectors at constant relative angle π is shown in fig. 2. Besides the ν_L component, which is clearly visible, the pattern exhibits a high frequency oscillation at $2\nu_L$. Its most pronounced effect is to give the low frequency peaks a "cusped" appearance.

The data may be analyzed most conveniently by Fourier transforming them into the frequency domain. This was accomplished by procedures analogous to those described previously [5,6], except that because of the extreme timing requirements, an analogue time-to-amplitude converter and a pulse-height analyzer were used in this case. The Fourier cosine transform of the autocorrelation function of the coincidence counting rate is shown in fig. 3. This curve is similar to an NMR line, but there is of course no rf field present.

The frequency $2\nu_L$ of 680 MHz shows up clearly in the Fourier transform function, as shown in fig. 3(b). This frequency is an order of magnitude higher than any observed previously by perturbed angular correlations.

C. Discussion

The parameters for ^{100}RhNi obtained by the two methods are given in Table 1. The slightly lower frequency obtained by the NMR/PAC method is attributable to inductive heating in this kind of experiment, since ν_L in ferromagnets decreases with increasing temperature. The linewidth of 4 MHz in the NMR/PAC experiment is considerably larger than that in the TDPAC experiment. This increase in linewidth is probably due to power broadening in the NMR/PAC case.

The minimum linewidth of 2.5 MHz obtained in the TDPAC experiment is still in excess of the natural linewidth of 0.94 MHz. Besides possible technical causes of this broadening, such as difficulties in the TDPAC measurement over very long time intervals, or broadening resulting from the rather involved data reduction procedures, or the finite length of the data sample autocorrelated, there are two possible inherent sources of the broadened line:

a) Inhomogeneous broadening. This kind of broadening is usual for conventional NMR linewidths in ferromagnetic metals and amounts typically to about $\Delta\nu_L/\nu_L = 10^{-2}$.

b) Relaxation broadening. This kind of broadening may be the consequence of eventual small damping of the correlation amplitude with time, because of possible nuclear spin-lattice relaxation interactions during the lifetime of the nuclear state.

Thus, it seems best, at least tentatively, to regard the 2.5 MHz linewidth as an upper limit for the two above-mentioned processes.

The value $H_{hf} = (-)\ 207.1(6)$ kG at 296 K is in good agreement with hyperfine field systematics [7]. A preliminary measurement giving $|H_{hf}\ (T = 473\ K)| = 164.3$ kG, a decrease of 21%, gives some indication that Rh in Ni might be exhibiting localized-moment behavior. Further study of $H_{hf}(T)$ is in progress.

It is a pleasure to thank Dr. E. Matthias for his continued interest in, and contributions to, the development of this field, Mr. George Gabor for his contributions to the development of both the NMR and the fast-timing apparatus, Mrs. Claudette Lederer and Mrs. Esther Schroeder for writing the computer programs used in our analysis, and Mrs. Winifred Heppler for carrying out and improving the sample preparation.

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Table 1

	NMR/PAC Method	TDPAC Method
Larmor frequency ν_L (MHz)	337.8(4)	339.5(3)
Linewidth $\Delta\nu_L$ (MHz)	4.0(4)	2.5(5)
Internal field (kG)[a]	(-) 206.0(6)	(-) 207.1(6)
Temperature (K)	$\geqslant$ 296	296.0(5)

[a] The errors include the error in the apparent g-factor, but not the error in the correction for the Knight shift of Rh in Rh metal. We have used the value of J. A. Seitchik, V. Jaccarino, and J. H. Wernick (Phys. Rev. 138 (1965) A148) of 0.43%, yielding g = 2.151. A recent redetermination of this shift as only 0.04% (T. H. Brown and P. J. Green, Phys. Letters 31A (1970) 148) would change the true value of g to 2.159, giving $|H_{hf}|$ values of 205.3 kG and 206.3 kG, respectively, for the NMR/PAC and TDPAC measurements.

Figure Captions

Fig. 1. NMR/PAC spectra for ^{100}RhNi: (a) ref. 1 (1966), (b) ref. 2 (1968), and (c) present results (1969). In (a) and (b) the wide and asymmetric absorption areas are attributed to the presence of Pd impurity in the Ni host. In (c), the low frequency component is no longer present and the effect is tripled to 6%.

Fig. 2. Part of a time-differential perturbed angular correlation spectrum for the 74.8 keV, 2^+ state of ^{100}Rh in Ni.

Fig. 3. Fourier cosine transform of the autocorrelation function of the coincidence counting rate showing (a) the Larmor resonance frequency ν_L = 339.5(3) MHz, yielding a hyperfine field $|H_{hf}|$ = 207.1(6) kG at T = 296 K, and (b) the high frequency component at $2\nu_L$ = 680 MHz.

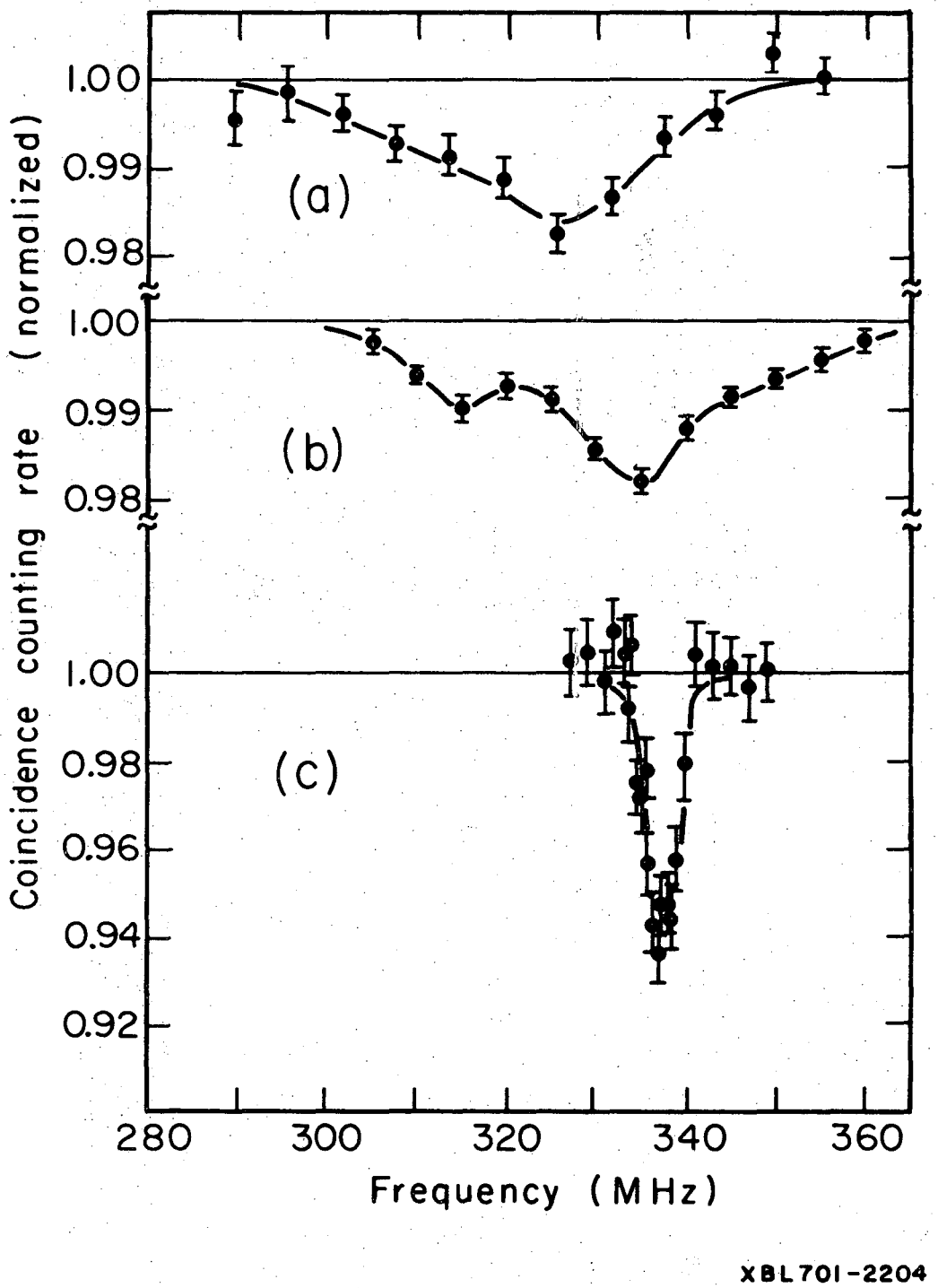
Coincidence counting rate (normalized)
1.00
0.99
0.98
(a)
1.00
0.99
0.98
(b)
1.00
0.98
0.96
0.94
0.92
(c)
280
300
320
340
360
Frequency (MHz)
XBL701-2204

Fig. 1

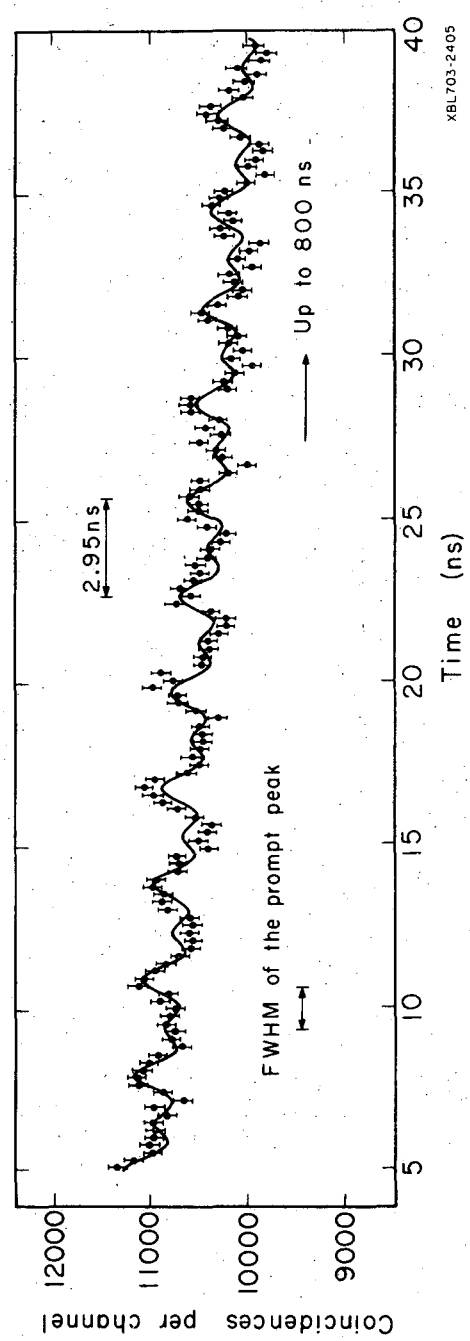
XBL703-2405
Coincidences per channel
12000
11000
10000
9000
5
10
15
20
25
30
35
40
Time (ns)
2.95ns
Up to 800 ns
FWHM of the prompt peak

Fig. 2

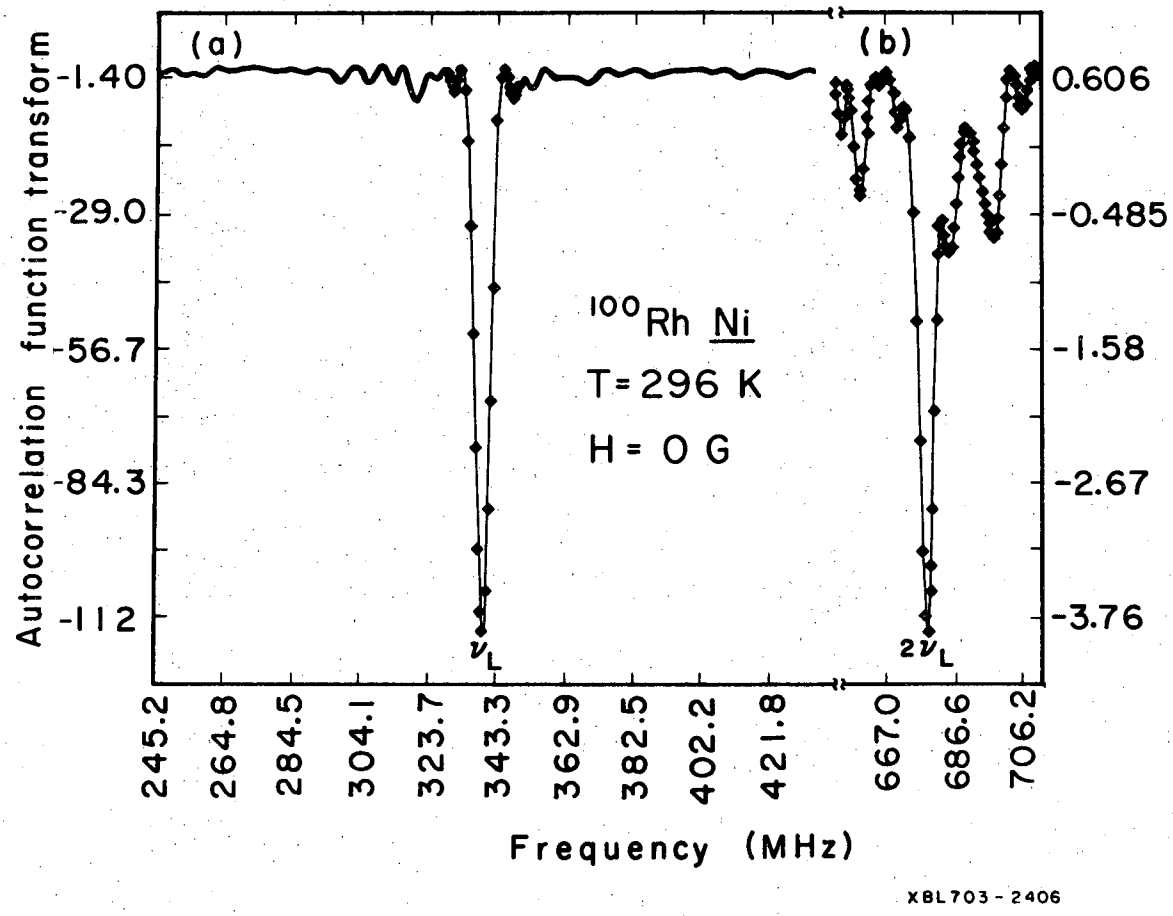
(a)
(b)
Autocorrelation function transform
-1.40
-29.0
-56.7
-84.3
-112
0.606
-0.485
-1.58
-2.67
-3.76
^{100}Rh Ni
T= 296 K
H = 0 G
ν_L
$2\nu_L$
245.2
264.8
284.5
304.1
323.7
343.3
362.9
382.5
402.2
421.8
667.0
686.6
706.2
Frequency (MHz)
XBL703-2406

Fig. 3

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