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DETECTION OF NUCLEAR MAGNETIC RESONANCE IN A 235 nSEC NUCLEAR STATE BY PERTURBED ANGULAR CORRELATIONS

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We have observed resonant rf absorption by 10^4 nuclei in the 74.8-keV excited state of Rh^{100} by combining nuclear magnetic resonance in a ferromagnet[1] with perturbed angular correlations (PAC) of the 84.0 - 74.8 keV γ-ray cascade.[2] The 4-day isotope Pd^{100}, which was diffused into a nickel foil, fed the cascade via electron-capture decay, maintaining an average of 0.006 nuclei in the 74.8-keV state during the experiment. A total of 2.8×10^6 coincidences were recorded by two 3-in x 3-in NaI(Tℓ) counters at 180°, while the sample was polarized in a field of 100 gauss along the counter axis. Perpendicular to the DC field an oscillating field of 3 gauss was stepped through the frequency range 289.5-355.5 MHz. Resonant absorption was detected by a drop in the coincidence rate centered about 322.5 MHz, as shown in Fig. 1.

The method of PAC has for sometime provided the only way to measure precession frequencies in short-lived nuclear states, with lifetimes in the range 10^{-11} sec $< \tau < 10^{-6}$ sec. Time-differential PAC,[3] and more recently the digital analysis method (DAPAC),[4] have raised the upper limit on lifetimes available to at least 10^{-3} sec and have greatly improved the accuracy possible with PAC. Still it is desirable to develop a method that combines the precision of NMR with the extremely high sensitivity of PAC in order to enjoy the advantages of both methods.

Abragam and Pound discussed the possibility of detecting resonant absorption in a short-lived nuclear state by observing the angular-correlation anisotropy as early as 1953.[5] They formulated the conditions for such an experiment:

(1) The splitting of the substates should be large compared to the natural nuclear linewidth; i.e., $\omega_L \tau_N >> 1$, where ω_L and τ_N are, respectively, the Larmor frequency and the nuclear lifetime.

(2) The rf field, H_1, must have sufficient amplitude to induce transitions in each nucleus in times of the order of the nuclear lifetime, $\gamma H_1 \sim \tau_N^{-1}$, where γ is the nuclear gyromagnetic ratio.

(3) Other time-dependent interactions should be minimal, so that the nuclear spin-correlation time τ_c is of the order of the lifetime, $\tau_c \sim \tau_N$.

These conditions, especially (2), turn out to be very stringent when applied to the PAC sources that have been studied, and the resonance experiment has generally not been considered feasible.

The effective rf field, H_1^{eff}, may be greatly enhanced by performing the resonance experiment on an atom embedded in a ferromagnetic lattice.[1] The enhancement arises through the hyperfine field H_{hf}, giving

$$2\ H_1^{eff} = (1+H_{hf}/H_o)H_1,$$

where H_o is the DC polarizing field. Thus, for the parameters that apply to our experiment, $H_o = 100G$ and $H_{hf} = 2 \cdot 10^5 G$, and the applied rf field is amplified by 10^3.

Rhodium-100 was chosen for this experiment because it is a well-understood case with a long lifetime and a large g-factor, and because hyperfine fields of the order of 10^5 gauss are expected for Rh dissolved

in the 3d ferromagnets. We selected a Ni lattice because nickel foil of the requisite thickness (10^{-4} cm = skin depth at 300 MHz) was available. In a recent study of the temperature-dependence of H_{hf} for Ru in Ni[6] it was found that the ratio $H_{hf}(295°K)/H_{hf}(0°K) = 0.80$; combining this with the value H_{hf}(Rh in Ni, 0°K) = -250 kG predicted from systematics,[7] we expected H_{hf}(Rh in Ni, 295°K) to have the magnitude 200 kG. This determined the frequency range selected for searching.

The essence of the experiment is very simple. The magnetization direction of the foil serves as the quantization axis and the propagation direction of the γ rays that were detected (the σ components, corresponding to $\Delta M_I = \pm 1$). For a coincidence to be recorded, σ components of the two transitions had to be registered within 1 μsec in the two counters, thus ensuring high probability (about 90%) that they were from the same nucleus. The coincidence counting rate under the experimental conditions varies with the angle θ between the propagation directions of γ_1 and γ_2 as $1 + 0.2\ P_2(\cos\theta)$ so with $\theta = 180°$ the coincidence rate is 20% larger than "average". This may be regarded as arising from the selection by the first counter, of a certain population distribution of nuclei in the 74.8-keV state that are candidates for coincidence (i.e., those that arrived in the 74.8-keV state via a σ transition). Radiation at the resonant radio frequency tends to randomize this population, thus lowering the coincidence counting rate. This is illustrated in Fig. 2, which shows rf transitions driving a nucleus that would have emitted a γ_2 σ component into a substate from which it may instead emit a π component.

Two features of Fig. 1 require comment. First, the absorption is small, of the order of 2% rather than 20%, and secondly the linewidth is

approximately 20 MHz, or about seven natural linewidths. These two features probably arise from the same cause, and were anticipated, because it has been found that the time-dependent correlation pattern of Ru^{99} in Ni loses coherence in $\sim 5 \times 10^{-8}$ sec. Such behavior could be the result of fluctuating local moments,[8] of quadrupole interaction, or simply of the existence of different magnetic sites. This point will be explored further. The fact that we can detect resonance in spite of this problem indicates that this type of experiment is feasible for shorter-lived states.

Comparison of the resonant frequency of 322.5 ± 3 MHz with the g-factor, +2.151 ± 0.004,[4] yields for H_{hf}(Rh in Ni, 290°K) a value of 197 ± 2 kG. This is in excellent agreement with the trends in this region, and quite close to the predicted value. The temperature dependence of H_{hf} is under further study. The sign of H_{hf} can be determined by radiating with circularly polarized rf radiation.

The combination of PAC and NMR should provide a method for investigating hyperfine interactions in short-lived excited nuclear states, in particular for levels with lifetimes between 10^{-6} and 1 sec, which are not accessible to other methods. For the longer lifetimes ferromagnetic enhancement should be unnecessary. For longer-lived states one has to produce the nuclear levels by a reaction process, which also provides larger anisotropies for the detection of the resonance and allows singles, rather than coincidence, detection. The limit of application will be set not by the nuclear lifetime, but by the relaxation times of the nuclei in their environments.

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8. We are indebted to A. M. Portis for pointing out this possible contribution to the linewidth.

FIGURE CAPTIONS

Fig. 1. Coincidence counting rate as a function of applied rf frequency, for the 84.0-74.8 keV γ-ray cascade of Rh^{100} in nickel.

Fig. 2. Energy-level diagram. Three levels of ^{100}Rh, of respective spins 1, 2, 1, are quantized in a hyperfine magnetic field parallel to the counter axis. Only the σ ($\Delta M = \pm 1$) components of dipole γ radiation, with distribution $\sim(1+\cos^2\theta)$, can strike counters at $\theta = 0°$ and $180°$. For a coincidence to be registered, σ components of both the 84.0-keV (γ_I) and the 74.8-keV (γ_2) transitions must be recorded within 1 μsec. We denote σ and π components by solid and dotted lines, and ΔM_I transitions within the 74.8-keV state by heavy lines. On the left a typical σ component of γ_1 is shown, with six of the many possible fates that may befall the nucleus in the intermediate (74.8-keV) state. Four of these paths end in σ transitions and are recorded as coincidence. The other two contribute to the dip in Fig. 1. Most of the possible components are omitted for clarity.

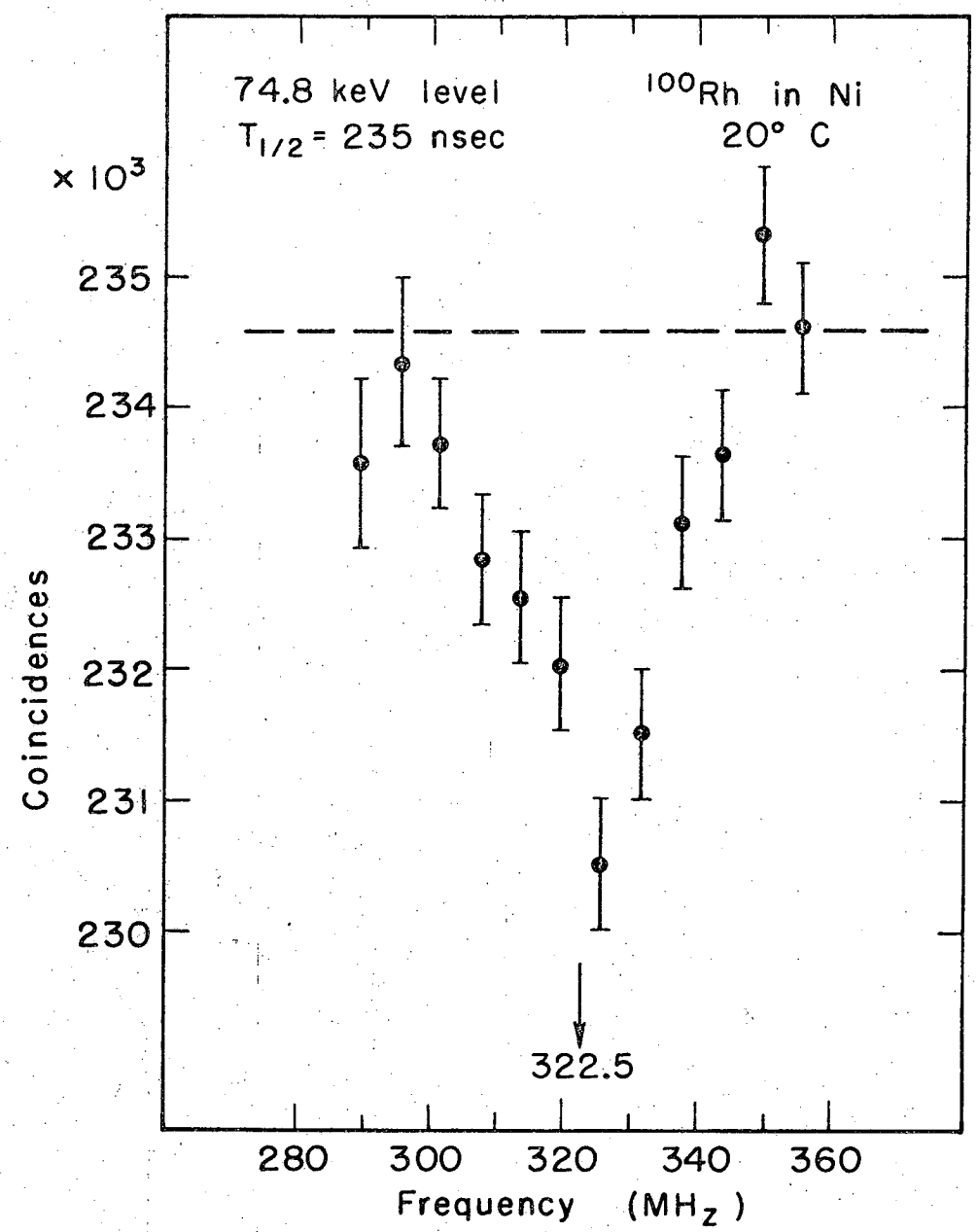

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Fig. 1

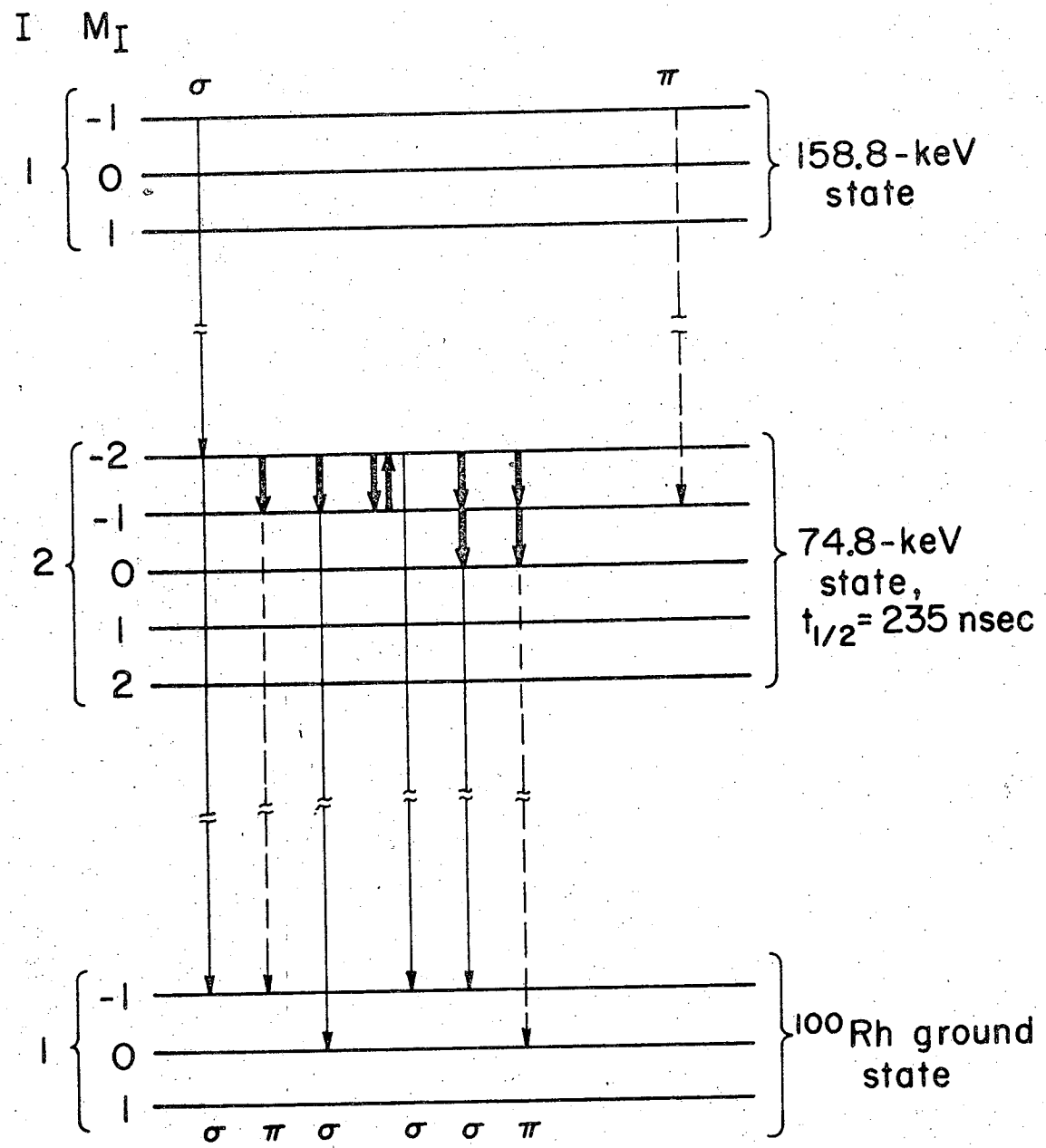
I
M_I
σ
π
1
-1
0
1
158.8-keV state
2
-2
-1
0
1
2
74.8-keV state, $t_{1/2}$= 235 nsec
1
-1
0
1
^{100}Rh ground state
σ π σ σ σ π
MUB-10402

Fig. 2

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