A Study of Multiple Quantum Transitions by Rotary Saturation of Nuclear Magnetic Resonance

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Multiple quantum transitions between the spin Zeeman levels of proton in water has been investigated by the rotary-saturation method in nuclear magnetic resonance. Experiments were carried out up to 3rd-order multiple quantum transitions. A quantitative analysis of the rotary-saturation data were accomplished by the use of the calculation of high-order quantum mechanical perturbation. The experimental results are in good agreement with the perturbation theory.

§ 1. Introduction

In the past decade, multiple quantum transitions of proton in water was investigated by molecular beem experiments[1]. The observation of multiple quantum transitions of spin system in liquids also performed by the rotary-saturation mehtod in nuclear magnetic resonance (which reported and called by Redfield[2]). Moreover, the rotary-saturation experiment is paticularly useful for the observation of multiple quantum transitions in soids. This novel method may be described in the following : When an alternating field H_1 is applied adiabatically on a spin system which subjected in a strong static field H_0, nuclear spin system will be parallel to H_{eff}, the effective field in the reference frame rotating with H_1. In this refernce frame, H_{eff} becomes the static field. Then if an audio-frequency (af) field H_a is applied along the static field H_0 (H_a having a component perpendicular to H_{eff}) and $\omega_a=\gamma H_{eff}$, one expects this af field to induce transitions in the spin system (where ω_a is the audio angular frequency and γ is the nuclear gyromagnetic ratio.).

We studied multiple quantum transitions of proton in water by the rotary-saturation method under the condition that proton spin system is subjected to the perturbing field in the short time compared with spin-spin and spin-lattice relaxation times. Then the problem is reduced to the following simple situation : Single magnetic dipole with nuclear spin quantum number $I=1/2$ is placed in time constant field H_{eff} in the reference frame. Application of a monocromatic radiation field H_a with angelar frequency $\omega_a=\gamma H_{eff}$ will induce the transition of a spin from a state $m=+1/2$ with energy ε_α to a state $m=-1/2$ with energy ε_β.

Then the condition

$$n\hbar\omega_a=\Delta\varepsilon \qquad n=1, 2, 3\cdots\cdots \tag{1}$$

is satisfied. ($\Delta\varepsilon=\varepsilon_\beta-\varepsilon_\alpha$) In the quantum mechanical description of electro-magnetic field, $\hbar\omega_a$ signifies a energy of a photon. The transition with $n=1$ corresponds to a single photon absorption, whereas the transition with n corresponds to a multi-photon absorption, i. e. multiple quantum transition. In the latter case, a single spin of proton is flipped by the absorption of n photons of radiation field.

A transition such that contributed by one more than single photon had discussed theoretically in 1929. At that time, M. Göppert-Mayer[3] showed that the transition probability is given by Dirac's radiation theory. The first experiment of multiple quantum process carried out by Hughes and Grabner[4], they observed a double quantum transition in $Rb^{87}F$ by the molecular beem electric resonance method.

We studied the multiple quantum transitions of proton spins in water by rotary-saturation method. The measurement of multiple quantum transitions is performed by obserbation of the free induction decay signal following soon after H_1 pulsed off. We treated the interaction between spin Zeeman levels and radiation field by the aid of Dirac's time dependent perturbation theory. Rotary-saturation spectra for $n=1, 2, 3$ were dscussed theoretically and the agreement between the experiments and the theory is quite well partially. We wish to study the multiple quantum transitions in solids in near future.

§ 2. Experimental procedure and apparatus

A. Experimental procedure

The technique of rotary saturation was developed by Redfield[2]. To perform a rotary-saturation experiment, it is necessary to add an audiro-frequency channel to a standard nuclear magnetic resonance appratus. The audio channel is used to probe spin systems in the rotating frame where the spins are quatized along an effective static field $H_{eff}=[H_1^2+(H_0-\omega/\gamma)^2]^{1/2}$. This rotating frame rotates about z-axis at angular frequency ω of radio-frequency (rf) field. Scince the af field H_a is created in the laboratory frame but viewed in the rotating frame, it is convenient to align H_a with the one axis shared by these two frames, the axis of the laboratory static field H_0. Redfield performed his original experiments using a steady-state apparatus. Our experiments have been utilised pulsed method.

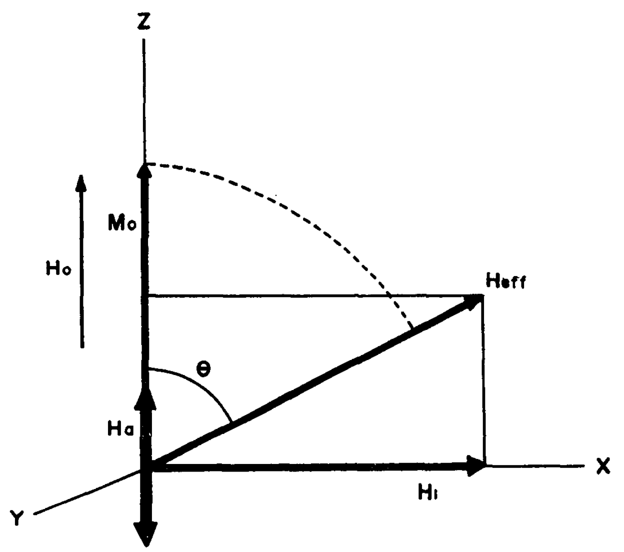

Fig. 1 Relationship of H_0, H_1, H_{eff} and H_a in the rotating coordinate system. Magnetization will follow to the effective field until making an arbitrary angle θ.

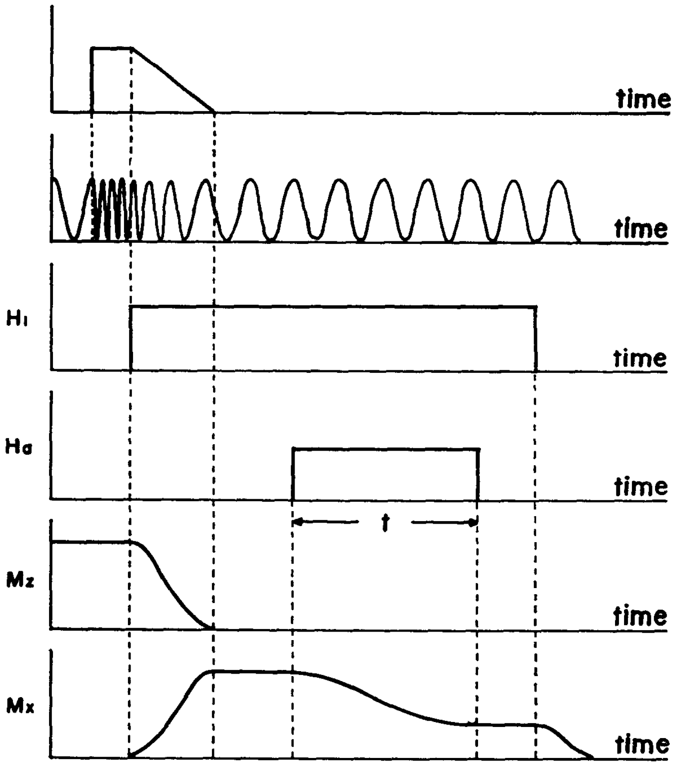

Fig. 2 Time dependence of magnetic fields (H_1 and H_a), the resulting magnetization components (M_z and M_x) and the frequency-modulated CW.

The spatial arrangements of the static field H_0, rf field H_1, af field H_a and magnetization M in the rotating reference frame are showen in Fig. 1. In this frame, a composition of H_0 and H_1 creates a resultant effective field $\boldsymbol{H}_{eff}=H_1\,\boldsymbol{i}+(H_0-\omega/\gamma)\boldsymbol{k}$. The pulse sequence used in our experiments is showen in Fig. 2.

Initially the spin system, consisting of the protons in water, is allowed to come to equilibrium in the laboratory frame in the presence of H_0 and H_1's zero, so that a magnetization vector points along H_0. Our first step involves turning on H_1 in such a manner that M is brought to point along H_{eff} in the rotating reference frame. If we simply turned on H_1, we would fail in our objective since, as showen in Ref. 5, M would precess around H_{eff} in the rotating frame, always remainning perpendicular to it and decaying in amplitude in a spin-spin relaxation time T_2 of spin system. In order to get M parallel to H_{eff} (called spin locking), we use the frequency modulation method[6]. The modulation width is about 137 KHz at about $\omega=15$ MHz, this frequency deviation corresponds to the field deviation 32 G for proton spins, which is quite large compared to the local field of the proton spins. Then this deviation $\Delta\omega$ slowly decreases to zero. By slowly, we mean taking a time of about 150 μsec., which is quite long compared to the precession period of the spins. During such a variation of ω, the magnetization becomes parallel to H_{eff} with suitable angle θ from z-axis, and then H_a is pulsed on. Then if resonance condition $n\omega_a=\gamma H_{eff}$ is satisfied, the component of H_a perpendicular to H_{eff} induced transitions of the spin system quantized along H_{eff} and M_x which is x component of the magnetization decreases. H_a is allowed to remain on for a variable time t short compared with H_1 on time and then pulsed off. The magnetization precesses about H_{eff}, and after the component of transverse to H_{eff} has had time to dephase, H_1 is pulsed off. The remaining magnetization precesses about H_0 and is detected by the free induction decay signal following soon after H_1 pulsed off. In such a manner, one can detect multiple quantum transitions by the reduction of the free induction decay signal. A curve is traced out by varying ω_a through resonant value $\omega_a = \gamma H_{eff}/n$. Measurements can be made to study the effect of varying H_1, H_a, t and the angle θ. All experiments were performed at room temperature.

B. Apparatus

The experiments were performed using the pulsed nuclear magnetic resonance apparatus with an audio channel shown in block diagram form in Fig. 3. The apparatus were essentially the same as that used by Franz and Slichter[7].

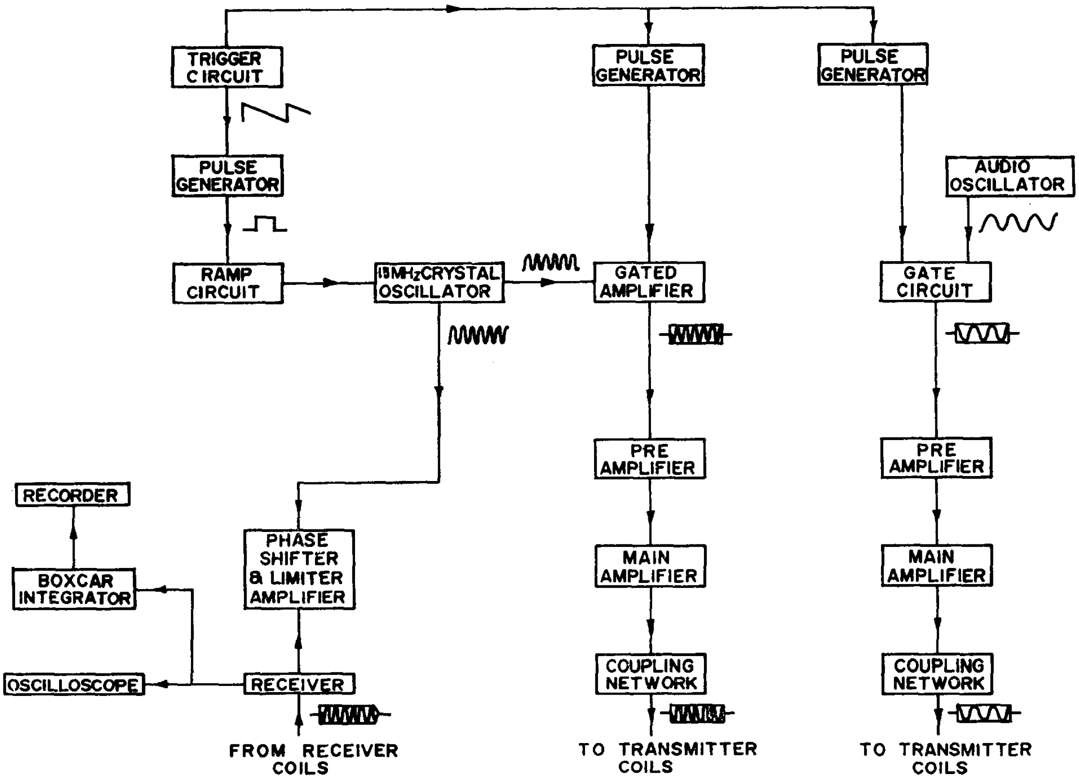

Fig. 3 Block diagram of rotary-saturation apparatus.

A frequency modulated continuous wave, 14.935 MHz signal, produced by a crystal oscillator, is fed into a gated amplifier. The pulsed rf output is then amplified by a preamplifier, class C power amplifier and passes through a coupling network to the transmitter coils. The signal produced by the precessing spins is picked up by the receiver coil, and passes through a low-noise preamplifier and high-gain rf amplifier to the detector. The detector was fed by a rf reference voltage differing slightly in frequency from 14.935 MHz, so that the output of the detector is the beet between the reference and the signal i. e. the envelope of the magnetization decay pattern. This is viewed directly with an oscilloscope and is also fed into a boxcar integrator which allows us to average over many pulses. The output of the boxcar goes directly to a X-Y recorder, so that a permanent record of the signal height is automatically produced.

The audio channel consists of an oscillator, a gate circuit, preamplifier and a power amlifier. The preamplifier and the power amplifier were used SONY 1130 type. The audio coils are wrapped on the outside of the rf probe.

The timing of the various pulses is controlled by a network of the several pulse generators. The repetition of the series of the pulses is defined on the wave form generator.

The rough estimation of the rf field strength was done by the viewing of the voltage across the transmitter coils on an oscilloscope. The accurate estimation of H_1 in gauss was obtained using a proton sample by measuring the length of 90° pulse and the equation $\gamma H_1 t = \pi/2$. The audio-amplitude would be calibrated in a completely analogous way, the main difference being that the pulse was applied in the second rotating frame. But in the experiments reported in this paper the caliblation was not done, since the fraction of the length of 90° pulse was large. In near future, we will perform the calibration of the audio-amplitude used in our experiments.

§ 3. Theory

A. Hamiltonian of systems

We start by assuming that a system of nuclear spins interact only with applied magnetic field and is isolated to each other and lattice. We consider that the applied magnetic field is an assembly of photons. Then the Hamiltonian of the systems in the rotating reference frame can be written as

$$\mathscr{H} = \mathscr{H}^S + \mathscr{H}^P + \mathscr{H}^1 \tag{2}$$

The first term gives the interaction of the spins with the effective static feld in the rotating frame, and by using the Pauli spin matrix $\boldsymbol{\sigma}$ we can write as

$$\mathscr{H}^S = -\mu\boldsymbol{\sigma} \cdot \boldsymbol{H}_{eff} \tag{3}$$

where $\mu = \gamma\hbar I$, is a magnetic moment of a spin and I is a spin quantum number,

$$\boldsymbol{H}_{eff} = H_1\, \boldsymbol{i} + [H_0 - \omega/\gamma]\boldsymbol{k} \tag{4}$$

The second term in Eq. (2) gives a Hamiltonian of a photon system, and by using a creation and an annihilation operator is described as

$$\mathscr{H}^P = \hbar\omega a^+ a, \qquad a^+ a = N \tag{5}$$

where N is an occupation number of photons in the one-photon state. These operators act on a function $|N\rangle$:

$$a|N\rangle = \sqrt{N}|N-1\rangle$$
$$a^+|N\rangle = \sqrt{N+1}\,|N+1\rangle \tag{6}$$

The state $|N\rangle$ represents a state constituted by N photons, that is, a transition from a state $|N-1\rangle$ to a state $|N\rangle$ corresponds to the diminution of one photon and the total energy of photon system decreases by amount of $\hbar\omega$.

The third term of Eq. (2) gives an energy of the interaction between a nuclear spin system and a photon system (i. e. applied magnetic field). We denote the vector potential of magnetic field by $\boldsymbol{A}$.

$$\mathscr{H}^I = -\mu\sigma \cdot rot\boldsymbol{A} \tag{7}$$

$$\boldsymbol{A} = \sqrt{\frac{2\pi\hbar c^2}{V\omega}}\,(ae^{i\boldsymbol{kr}} + a^+e^{-i\boldsymbol{kr}})\,\boldsymbol{e} \tag{8}$$

$$|\boldsymbol{k}| = \omega/c, \qquad |\boldsymbol{e}| = 1, \qquad \boldsymbol{ke} = 0$$

where $\boldsymbol{k}$ is a wave vector, $\boldsymbol{e}$ is a polarization vector and V is a volume. Then

$$rot\boldsymbol{A} = i\sqrt{\frac{2\pi\hbar\omega}{V}}\,(ae^{i\boldsymbol{kr}} - a^+e^{-i\boldsymbol{kr}})\,\boldsymbol{b} \tag{9}$$

$$\boldsymbol{b} = c/\omega[\boldsymbol{ke}], \qquad |\boldsymbol{b}| = 1$$

We have, by using Eqs. (7), (9) and the dipolar approximation ($\boldsymbol{kr} << 1$)

$$\mathscr{H}^I = -i\mu\sqrt{\frac{2\pi\hbar\omega}{V}}\,(a - a^+)\,\sigma\boldsymbol{b} \tag{10}$$

B. Derivation of the expession of transition probability from the perturbation theory

The treatment of a system for which the Hamiltonian depends on the time was given by Dirac[8] by using the method of the time-dependent perturbation theory. We start from the assumption that

$$\mathscr{H} = \mathscr{H}^0 + \mathscr{H}^I, \qquad \mathscr{H}^0 u_m = E_m u_m \tag{11}$$

where the unperturbed Hamiltonian $\mathscr{H}^0$ can be solved for its normalized eigenfunctions u_m and its enery eigenvalues E_m, and the perturbation $\mathscr{H}^I$ is small. We consider to solve the time-dependent Schroginger equation

$$i\hbar\frac{\partial\Phi}{\partial t} = \mathscr{H}\Phi \tag{12}$$

The solution of Eq. (12) is given as the expansion of the wave equation starts from $\Phi(0) = u_l$,

$$\Phi(t) = \Sigma_m\, a_{ml}(t)\, u_m e^{-i\omega_m t} \tag{13}$$

where $\omega_m = E_m(0)/\hbar$, $a_{ml}(t)$ is a probability amplitude finding a system in one of the eigenstate $u_m \exp\{-iE_m(0)t/\hbar\}$ at the time t.

The substituting Eqs. (11) and (13) into Eq. (12) yields the set of equations

$$\frac{da_{ml}(t)}{dt} = \frac{1}{i\hbar}\sum_k \mathscr{H}^I_{mk}(t)\, a_{kl}(t) \tag{14}$$

where

$$\mathscr{H}^I_{mk}(t) = \mathscr{H}^I_{mk}(t)\, e^{-\frac{i}{\hbar}(E_k - E_m)t}$$

In general Eq. (14) is an infinite simultaneous equations, but in the case which the number of eigenstate of $\mathscr{H}^0$ is finite, under the suitable initial condition can be resolved by the method of successive approximation. The solution for n-th order is given as follows :

$$a_{ml}^{(n)}(t) = \delta_{ml} + \sum_{k=1}^{n}\left(\frac{1}{i\hbar}\right)\int_{o}^{t} dt_1 \int_{0}^{t_1} dt_2 \cdots\cdots \int_{0}^{t_{k-1}} dt_k \mathscr{H}^{I}_{mr_1}(t_1)\, \mathscr{H}^{I}_{mr_2}(t_2) \cdots\cdots \mathscr{H}^{I}_{mr_k}(t_k) \tag{15}$$

Now we consider the situation that the interaction time t is variable but $\mathscr{H}^I$ is time-independent, and then Eq. (15) is rewritten as follows

$$a_{ml}^{(n)}(t) = \delta_{ml} + \sum_{k-1}^{n}\left(\frac{1}{i\hbar}\right)^{k} \sum_{r_1}\cdots\sum_{r_{k-1}} \mathscr{H}^{I(k)}_{ml} \int_{0}^{t} dt_1 \int_{0}^{t_1} dt_2 \cdots \int_{0}^{t_{k-1}} dt_k e^{i\alpha(t_1,\cdots,t_k)} \tag{16}$$

where

$$\mathscr{H}^{I(k)}_{ml} = \mathscr{H}^{I}_{mr_1} \mathscr{H}^{I}_{r_1r_2} \cdots \mathscr{H}^{I}_{r_{k-1}l}$$

$$\alpha(t_1, t_2, \cdots, t_k) = \frac{1}{\hbar}(E_m - E_{r_1})t_1 + \frac{1}{\hbar}(E_{r_1} - E_{r_2})t_2 + \cdots + \frac{1}{\hbar}(E_{r_{k-1}} - E_l)t_k$$

E_{v_i} is a eigenvalue of operator $\mathscr{H}^0$. Integrating Eq. (16) gives

$$a_{ml}^{(n)}(t) = \delta_{ml} + \sum_{k=1}^{n} \sum_{r_1}\cdots\sum_{r_{k-1}} \mathscr{H}^{I(k)}_{ml} \sum_{\rho} \frac{e^{\frac{i}{\hbar}(E_m - E_\rho)t}}{(E_\rho - E_m)\prod_{\alpha}'(E_\rho - E_\sigma)} \tag{17}$$

$$\rho = l, r_1, \cdots, r_{k-1}, \qquad \sigma = l, r_1, \cdots, r_{k-1} \neq \rho$$

The transition probability from a state $|l>$ to a state $|m>$ after the time interval t is given by

$$p_{l\to m}(t) = |a_{ml}(t)|^2 \tag{18}$$

C. Application of the perturbation theory to $I = 1/2$ spin system

We consider an application of the results of perturbation theory to a spin system constituted with $I = 1/2$ spins in an alternating magnetic field. The denominator in Eq. (17) is a difference between eigenvalues of an operator $\mathscr{H}^0$. The energy of total systems corresponding to a eigenstate $|r\rangle$ is

$$E = \hbar\omega N_r + \varepsilon_r \tag{19}$$

wheere N_r is the number of photons in the state charactelized by r, and ε_r is the energy of the spin system. For two states $|r\rangle$ and $|r'\rangle$

$$\hbar\omega(N_r - N_{r_1}) = \varepsilon_{r_1} - \varepsilon \tag{20}$$

Finding from Eq. (10) $\mathscr{H}^I$ is a linear function of operators a and a^+. Therefore the condition $|N_{r_1} - N_r| = 1$ is necessary for the matrix element of $\mathscr{H}^I$ being not zero. For $\mathscr{H}^{I(k)}_{ml}$, the k-th order product of $\mathscr{H}^I$, in Eq. (17) following condition must be satisfied.

$$|N_m - N_{r_1}| = 1, \quad |N_{r_1} - N_{r_2}| = 1, \cdots\cdots, |N_{r_{k-1}} - N_l| = 1$$

N_r is an occupation number of photons in a medium state $|r\rangle$ and the occupation difference between the initial state and the final state, $|N_l - N_m| = k$, gives the order of multiple quantum transitions. Then the condition for which one can observe the resonance of n-th order multiple quantum transitions is $N_l - N_m = n$, where n is over any positive integers. Since we consider the transition from the initial state $|l\rangle$ to the final state $|m\rangle$ of spin system by absorption of energy from photon system, Eq. (17) is rewritten by using Eq. (19) as follows:

$$a_{ml}^{(n)}(t) = -\frac{e^{\frac{i}{\hbar}[\hbar\omega(N_m - N_l) + \varepsilon_m - \varepsilon_l]t} - 1}{\hbar\omega(N_m - N_l) + \varepsilon_m - \varepsilon_l} k_{ml}^{(n)} \tag{21}$$

$$k_{ml}^{(n)} = \sum_{r_1} \cdots \sum_{r_{n-1}} \frac{\mathscr{H}^I_{mr_1} \mathscr{H}^I_{r_1 r_2} \cdots \mathscr{H}^I_{r_{k-1} l}}{\prod_v [\hbar\omega(N_l - N_{r_v}) + \varepsilon_l - \varepsilon_{r_v}]} \tag{22}$$

$$v = 1, 2, \cdots\cdots, n-1$$

Then we obtain the transition probability of n-quanta transition

$$p_{l\to m}^{(n)}(t) = |a_{ml}^{(n)}(t)|^2 = \left(\frac{\sin \frac{t}{2\hbar} [\hbar\omega(N_m - N_l) + \varepsilon_m - \varepsilon_l]}{\frac{1}{2}[\hbar\omega(N_m - N_l) + \varepsilon_m - \varepsilon_l]} \right)^2 |k_{ml}^{(n)}|^2 \tag{23}$$

Since up to the present we have used the index $l, m, \cdots\cdots$ for specifying the total system, hereafter we will use the additional index $\alpha, \beta, \cdots\cdots$ as the quantum numbers to specify the spin system. We assume that the total system starts from the state $|l\rangle = |N, \alpha\rangle$ and goes to the state $|m\rangle = N', \beta\rangle$ finally, so that, the transition of the spin system is induced from a state $|\alpha\rangle$ to a state $|\beta\rangle$ by absorbing $n(=N-N')$ photons in the photon system.

The energy of interaction between a spin system and a photon system is given by Eq. (6) and (10), since we treat absorption only

$$\langle N-1, \beta | \mathscr{H}^I | N, \alpha \rangle = -i\mu \sqrt{\frac{2\pi \hbar \omega N}{V}} (\sigma \boldsymbol{b})_{\beta\alpha} \tag{24}$$

Then the probability that an interaction $\mathscr{H}^I$ induces a transition from a state $|\alpha\rangle$ to a state $|\beta\rangle$ is given by substituting Eq. (24) into Eq. (23).

$$p_{\alpha\to\beta}^{(n)}(t) = \left(\frac{\sin \frac{t}{2\hbar} [n\hbar\omega - \Delta\varepsilon]}{\frac{1}{2}[n\hbar\omega - \Delta\varepsilon]} \right)^2 |k_{\beta\alpha}^{(n)}|^2 \tag{25}$$

$$|k_{\beta\alpha}^{(n)}|^2 = \left(2\pi\mu^2 \frac{\hbar\omega N}{V}\right)^n \left| \sum_{\alpha_1} \cdots \sum_{\alpha_{n-1}} \frac{(\sigma b)_{\beta\alpha_1} \cdots (\sigma b)_{\alpha_{n-1}\alpha}}{\prod [\hbar\omega(n-v) - |\varepsilon_\alpha - \varepsilon_{\alpha_v}|]} \right|^2 \tag{26}$$

$$v = 1, 2, \cdots\cdots, n-1$$

where $\Delta\varepsilon = \varepsilon_\beta - \varepsilon_\alpha$. Eq. (25) represents the transition probability of spin system following the absorption of n-quanta from an alternating magnetic field.

We denote the energy density of radiation, $\hbar\omega N/V$ in Eq. (26), by a symbol ρ, so that Eq. (26) becomes

$$|k_{\beta\alpha}^{(n)}|^2 = (2\pi\mu^2\rho)^n \left| \sum_{\alpha_1}\cdots\sum_{\alpha_{n-1}} \frac{(\sigma b)_{\beta\alpha_1}\cdots(\sigma b)_{\alpha_{n-1}\alpha}}{\prod_{v}[\hbar\omega(n-v) - |\varepsilon_\alpha - \varepsilon_{\alpha_v}|]} \right|^2 \tag{27}$$

When $n\hbar\omega = \Delta\varepsilon$, Eq. (25) becomes

$$p_{\alpha\to\beta}^{(n)}(t) = \left(\frac{t}{\hbar}\right)^2 |k_{\beta\alpha}^{(n)}|^2 \tag{28}$$

While $\boldsymbol{b}$ is a unit vector parallel to the axis of radiation coil, the scalor product of $\boldsymbol{\sigma}$ and $\boldsymbol{b}$ yields

$$(\sigma\boldsymbol{b}) = \begin{pmatrix} \cos\theta & e^{-i\phi}\sin\theta \\ e^{i\phi}\sin\theta & -\cos\theta \end{pmatrix} \tag{29}$$

where θ is an angle between H_{eff} and $\boldsymbol{b}$, and ϕ is an azimuth angle. By substituting Eq. (29) and the relation $\hbar\omega n = \Delta\varepsilon$ into Eq. (27)

$$|k_{\beta\alpha}^{(n)}|^2 = \left[\frac{(\mu\sqrt{2\pi\rho})^n}{(\Delta\varepsilon)^{n-1}} f_n(\theta)\right]^2 \tag{30}$$

where

$$f_1(\theta) = \sin\theta \tag{31a}$$

$$f_2(\theta) = 4\sin\theta\cos\theta \tag{31b}$$

$$f_3(\theta) = \frac{9}{4}\sin\theta(9\cos^2\theta - 1) \tag{31c}$$

Eq. (28), (30) and (31) are compared with the experimental results.

§ 4. Experimental results and discussion

In this section the experimental results will be presented and compared with the perturbation theory developed in Sec. 3. Figure 4 shows a typical rotary-saturation spectrum. The fraction of magnetization remaining after audio pulses is shown as a function of audio frequency $\omega_a/2\pi$ in this figure. Experimental conditions are $\tau=3$ $m$sec., $H_{eff}=2.5$ G, $H_a=30$ V and $\theta=79°$. A peak with 10.5 KHz corresponds to a single photon absorption. Finding from the relation $H_{eff}=\omega_a/\gamma$, the corresponding field strength to this peak is 2.5 G and is well agreement with the field strength 2.7 G from 90° pulse method. As a prediction of the the theory we can see a considerable destruction of magnetization at nearly one-half and one-third values of 10.5 KHz. These peaks correspond to a two-quanta and a three-puanta transitions in a spin system. Figure 5 shows also a complete rotary-saturation spectrum, where experimental conditions are $\tau=4$ $m$sec., $H_{eff}=2.4$ G, $H_a=30$ V and $\theta=79°$. This spetrum also shows the results predicted by the theory.

Now we will consider the time dpendency and the field strength dependency of radiation on the muliple quantum transition signals. To do so, we will find the tsansition probability per second, $p_0^{(n)}(t)$. We substitute the relations $\Delta\varepsilon=\gamma\hbar H_{eff}$, $\mu=\gamma\hbar/2$ and $\rho=H_a/8\pi$ into Eq. (30), and then using Eq. (28)

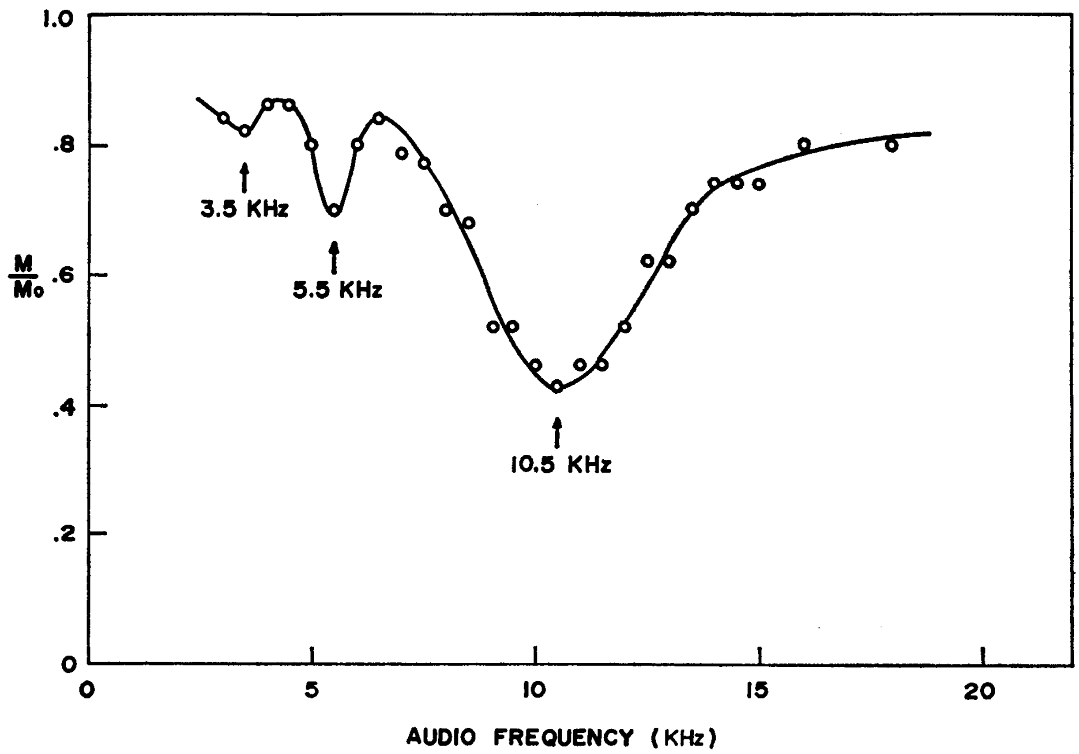

Fig. 4 The rotary-saturation spectrum showing one, two and three-quanta transitions.

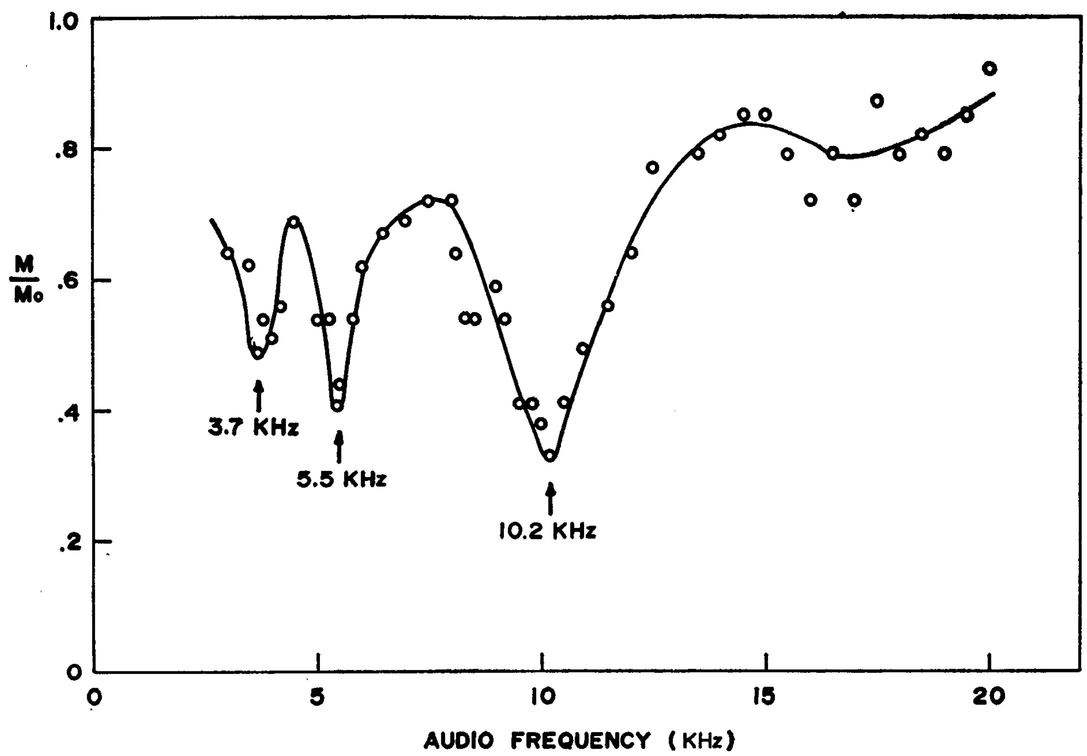

Fig. 5 The rotary-saturation spectrum.

$$p_0^{(n)}(t) = [\frac{\gamma(H_a/4)^n}{(H_{eff})^{n-1}} f_n(\theta)]^2 t \tag{32}$$

Since the destruction rate of magnetization is propotinal to the transition probabilyty per second. Using Eq. (32), the equation of the fraction of magnetization becomes

$$\frac{M}{M_0} = \exp\{-\frac{1}{2}[\frac{\gamma(H_a/4)^n}{(H_{eff})^{n-1}} f_n(\theta)]^2 t^2\} \tag{33}$$

We turn first to a discussion of the time dependency of radiation on the transition probability. In Eq. (33), putting H_{eff} and H_a to be constant

$$\frac{M}{M_0} = \exp\{-[g(n) f_n(\theta)]^2 t^2\} \tag{34}$$

where

$$g(n) = \frac{\gamma(H_a/4)^n}{\sqrt{2}\,(H_{eff})^{n-1}}$$

The fraction of magnetization remaining after an audio-frepuency pulse for $n=1$ (single quantum transition) is shown as a function of the radiation time t in figure 6, with experimental

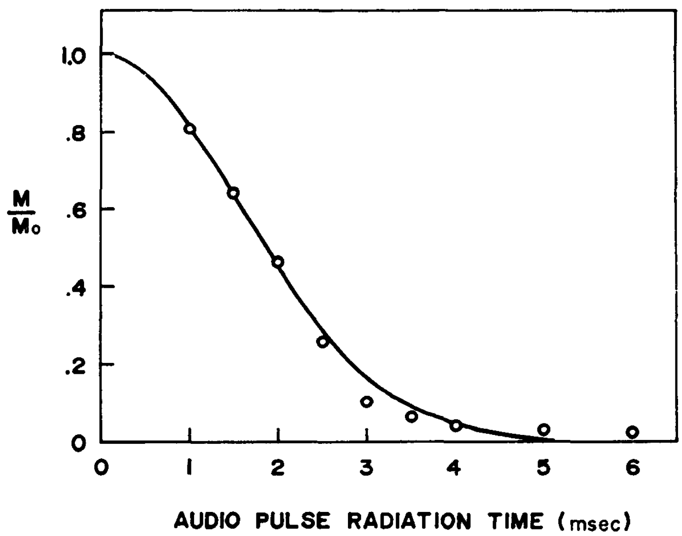

Fig. 6 Dependence of the fraction of magnetization on a radiation time of the audio-frequency pulse for single-quantum transition. The circles represent the obseved values. The line is drawn by using Eq. (34) with an adjustable value of H_a.

conditions of $H_{eff}=2.4$ G, $H_a=30$V and $\theta=80°$. In this figure, the circles represent the observed values and the line is plots of the theoretical Eq. (34) with adjustable value of H_a. Finding from this figure, the observd values agree with the theoretical values.

Next, we turn to a discusion of the field strength dependency of radistion. In this case, by putting t and H_{eff} to be constant, Eq. (33) is rewritten as

$$\frac{M}{M_0} = \exp\{-[g'(n) f_n(\theta)]^2 H_a^{2n}\} \tag{35}$$

where

$$g'(n) = \frac{\gamma\tau}{(4^n)\sqrt{2}\,(H_{eff})^{n-1}}$$

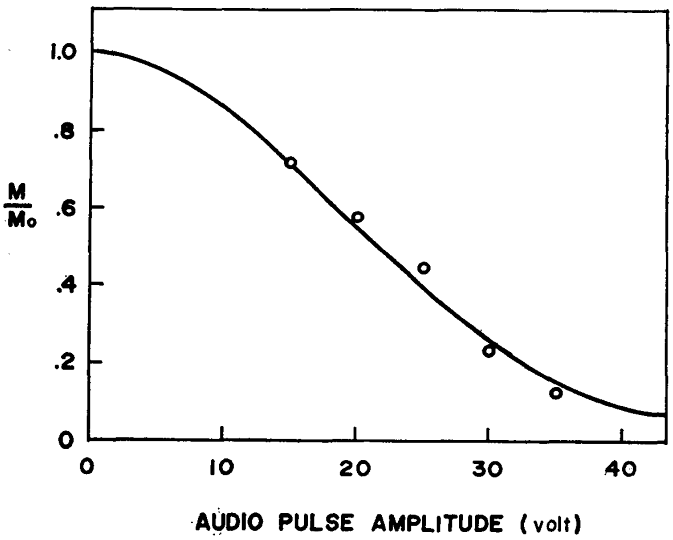

Fig. 7 Dependence of the fraction of magnetization on an audio-frepuency pulse strength for sigle-quantum transition. The circles represent the observed values. The line is drawn by using Ep. (35) with one adjustable value.

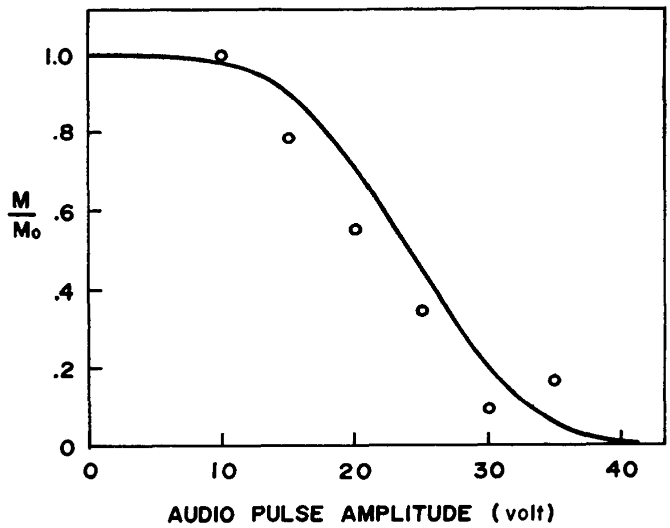

Fig. 8 Dependence of the fraction of magnetization on an audio-frequency pulse strength for two-puanta transitions. The circles represent the observed values. The line is drawn by using Eq. (35) with one adjustable value.

Figure 7 represents the fraction of magnetization remaining after an audio-frepuency pulse for $n=1$ (single-quantum transition) as a function of audio pulse length for various values of H_a. Experimental conditions are $H_{eff}=2.4$ G and $\theta=80°$. The circles are experimental values and the line is drawn by using the theoretical Eq. (35) with one adjustable value. The close agreement between the observd values and the theoretical curve was obtained. The results for the case of $n=2$ (two-quanta transitions) is shown in Fig. (8). Experimental conditions are $H_{eff}=2.4$ G and $\theta=85°$. In this case, the observed values are not well agreement with the theoretical curve.

The sudject of the study reported in this paper was liquid. We will study the multiple quantum transitions in solids systematically in near future.

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