ductance with the sharp boundary condition,[2] namely, exact matching of the wave functions, rather than with the WKB approximation. His result eliminates the cusp at $eV = F_p$ and thus is closer to the experimental result. However, quantitative discrepancy between the theory and the experiment in the negative-resistance region is believed to be due to the simplification employed here. See Ref. 3.

[10]J. A. Kafalas, R. F. Brebrick, and A. J. Strauss, Appl. Phys. Letters 4, 93 (1964); B. G. Bylander, J. R. Dixon, H. R. Riedl, and R. B. Schoolar, Phys. Rev. 138, A864 (1965); B. A. Efimova, V. I. Kaidanov, B. Ya. Moizhes, and I. A. Chernik, Fiz. Tverd. Tela 7, 2524 (1965) [translation: Soviet Phys.—Solid State 7, 2032 (1966)].

[11]The present effect would not be important as long as the applied voltage is kept low in comparison with the barrier height as seen in L. Esaki and P. J. Stiles, Phys. Rev. Letters 14, 902 (1965); 16, 574 (1966). In such cases, one can assume the tunneling exponent λ to be virtually constant in Eq. (1), as was done in Ref. 2.

SPIN-CLUSTER RESONANCE IN $CoCl_2 \cdot 2H_2O$

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A new type of magnetic resonance has been observed in $CoCl_2 \cdot 2H_2O$ at low temperatures. This compound is antiferromagnetic below 17.2°K with an antiferromagnetic arrangement of ferromagnetic linear chains. Exchange interactions among chains are antiferromagnetic while a strong ferromagnetic exchange interaction exists in the chain and an anisotropy favoring the b axis is so strong that the spin system can be looked at nearly as a bundle of Ising spin chains.[1,2] When an external magnetic field is applied along the b axis, the magnetization increases stepwise with increasing field[3,4] and, as was preliminarily reported by the present authors, curious resonances are observed[5] near the critical fields H_{c1} and H_{c2} (see Fig. 1). These resonances, which have been excited with microwaves of 24-70 Gc/sec in a pulsed magnetic field up to 50 kOe, are very different from usual ferromagnetic or antiferromagnetic resonances. For instance, the absorption intensity is weak and decreases rapidly with decreasing temperature from 4.2 to 1.5°K. We have now been able to analyze the detailed structure of the resonances and find that these absorptions come from a spin-cluster resonance with the selection rule $\Delta m = \pm 1$, where m is the number of spins in a short-range-order spin cluster directed opposite to the majority spins in a ferromagnetic chain.

Let us consider an Ising spin chain coupled by a ferromagnetic exchange interaction J_0, with an external magnetic field H_0 parallel to the spin easy axis (the b axis in $CoCl_2 \cdot 2H_2O$) which we take as the z axis. There is no phase transition in such a system, but almost all spins point along H_0 for a sufficiently strong field at low temperatures. However, some spins point toward $-z$ so as to minimize the free energy. Examples of spin clusters are shown in Fig. 2(a). Neglecting dipolar interactions, the resonance condition for the allowed transitions $\Delta m = \pm 1$ is simply $\omega/\gamma = H_0$. It should be emphasized that the resonance frequency for a spin-cluster transition does not depend on J_0 as there is no change in exchange ener-

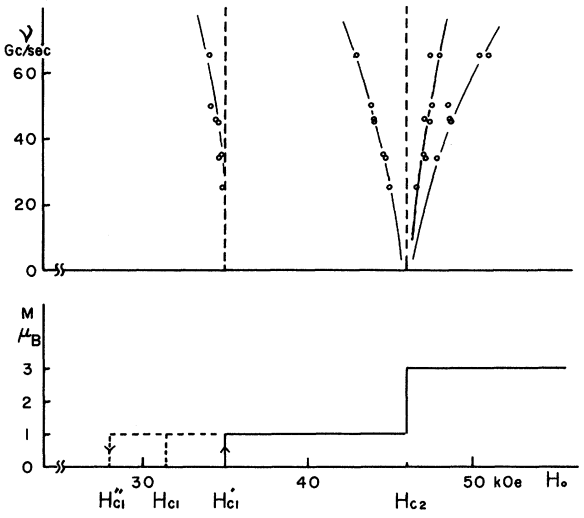

FIG. 1. Frequency-field diagram of the resonance points and corresponding changes of the magnetization ($H_0 \parallel b$ axis). H_{c1} is the thermodynamic critical field for the lower transition, but because of hysteresis, this transition is observed at H_{c1}' on increasing the field, and at H_{c2}'' when the field is decreased. Resonances near the lower transition field can only be observed in an increasing magnetic field, near H_{c1}'.

gy when a terminal spin of a cluster reverses direction. Now consider the absorption intensities as a function of H_0 and T. The absorption is proportional to the population differences between clusters of size m and $m+1$, summed over all clusters. This sum is just equal to the number of $m=1$ clusters, i.e., "clusters" of a single spin. This fact is also expressed in other words: For all $m \neq 1$, the transition probabilities for $\Delta m = 1$ and $\Delta m = -1$ are equal, so that there is no net absorption. But for single-spin clusters, while the transition $m=1 \rightarrow m=2$ can accompany absorption of a photon, the transition $\Delta m = -1$ is forbidden as the annihilation of the cluster liberates exchange energy $2J_0$ so that the resonance condition shifts far from the microwave region. Hereafter, the number of $m=1$ spin clusters is called the effective spin number, written as n_e.

To describe this qualitatively, assume a looped chain of N particles each having spin S. The internal energy is

$$U = -\sum_{i \neq j} 2J_0 S_i S_j - \sum_i g\mu_B H_0 S_i$$
$$= -(N-4n_c)J_0/2 - (N-2n_s)g\mu_B H_0/2, \quad J_0 > 0, \quad (1)$$

where S_i and S_j represent Ising spins, being equal to either $+\frac{1}{2}$ or $-\frac{1}{2}$ along the z axis, n_s is the total number of magnetic moments pointed in the $-z$ direction, and n_c is the number of spin clusters. The partition function of an Ising linear chain is

$$Z = [\exp(J_0/2kT)\{\cosh(g\mu_B H_0 S/kT) + R\}]^N, \quad (2)$$

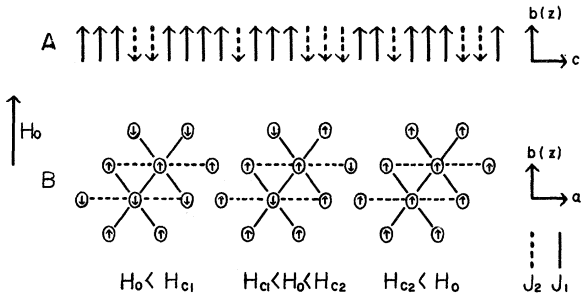

FIG. 2. Several short-range-order spin clusters randomly distributed in a chain are shown by dotted arrows in (a), and spin structures corresponding to the three phases are shown in (b). Nearest- and next-nearest-neighbor exchange couplings are indicated by solid and dashed lines, respectively, in (b).

where the notation

$$R = \{\sinh^2(g\mu_B H_0 S/kT) + \exp(-2J_0/kT)\}^{1/2},$$

is used for simplicity.

The free energy F, magnetization M, and internal energy U can be calculated from Eq. (2) as

$$F = -kT \ln Z$$
$$= -NJ_0/2 - NkT \ln\{\cosh(g\mu_B H_0 S/kT) + R\}, \quad (3)$$

$$M = -\partial F/\partial H_0 = Ng\mu_B S \sinh(g\mu_B H_0 S/kT)/R, \quad (4)$$

$$U = \partial(F/T)/\partial T^{-1}$$
$$= -NJ_0/2 - MH_0 + NJ_0$$
$$\times \exp(-2J_0/kT)/\{\cosh(g\mu_B H_0 S/kT) + R\}R. \quad (5)$$

Comparing Eq. (4) with the saturation magnetization $M_0 = Ng\mu_B S$,

$$n_s/N = (M_0 - M)/2M_0$$
$$= \{1 - \sinh(g\mu_B H_0 S/kT)/R\}/2. \quad (6)$$

n_c/N is obtained by comparing Eq. (1) with (5),

$$n_c/N$$
$$= \exp(-2J_0/kT)/2\{\cosh(g\mu_B H_0 S/kT) + R\}R. \quad (7)$$

The resonance effective spin number n_e is then obtained by statistical calculation[6] and the result is

$$n_e = n_c^2/n_s. \quad (8)$$

The maximum value of n_e lies near $H_0 = 0$ and it decreases rapidly with increasing field.

Now we discuss the resonances observed in $CoCl_2 \cdot 2H_2O$. Spin structures corresponding with three phases are shown in Fig. 2(b). It is assumed that for the spin-cluster resonance, effective fields acting on each cluster are the sum of H_0 and the exchange fields H_1 and H_2 which come from the surrounding chains. H_1 and H_2 are related to the exchange constants by $2|J_1|S/g\mu_B$ and $2|J_2|S/g\mu_B$, respectively. The critical fields can be written in terms of H_1 and H_2 as[3,4]

$$H_{c1} = 4H_1 - 4H_2 = 32 \text{ kOe},$$
$$H_{c2} = 4H_1 + 2H_2 = 46 \text{ kOe}, \quad (9)$$

Following the treatment of impurity-spin resonance in $FeCl_2$,[7] spin-cluster resonance conditions corresponding to the three phases are obtained as follows.

(1) $H_0 < H_{c1}$: There are two resonance branches (I) and (II) corresponding to transitions in chains pointing up and down, and the resonance conditions are given by

$$\text{(I)}\quad \omega/\gamma = 4H_1 - 2H_2 + H_0, \tag{10}$$

$$\text{(II)}\quad \omega/\gamma = 4H_1 - 2H_2 - H_0. \tag{11}$$

(2) $H_{c1} < H_0 < H_{c2}$: The exchange field for clusters in negative chains is $4H_1 + 2H_2$ but it is zero for positive chains:

$$\text{(III)}\quad \omega/\gamma = 4H_1 + 2H_2 - H_0, \tag{12}$$

$$\text{(IV)}\quad \omega/\gamma = H_0. \tag{13}$$

(3) $H_{c2} < H_0$: Similarly, the resonance condition is derived as

$$\text{(V)}\quad \omega/\gamma = H_0 - (4H_1 + 2H_2). \tag{14}$$

The frequency-field diagrams of branches (I)-(V) are shown in Fig. 3 using a g factor $g_b = 7.3$.[3] Since the absorption intensity depends on n_e, the resonance peaks determined by Eqs. (10)-(14) will be modified by linewidth effects. Taking a linewidth of 5 kOe, and with

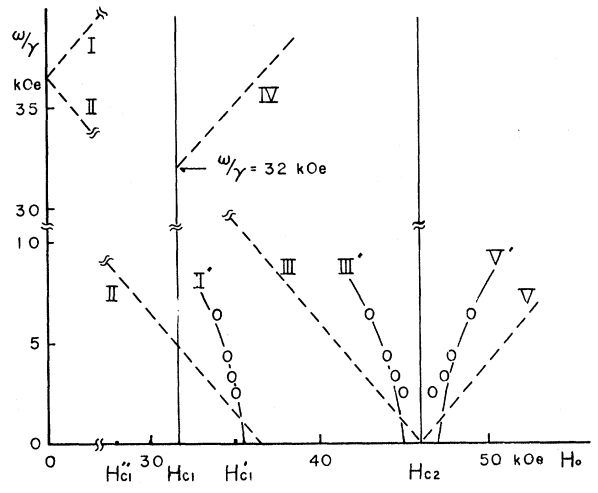

FIG. 3. Frequency-field diagram of branches (I)-(V) are shown by broken lines. These curves are modified by consideration of n_e to yield the final theoretical branches (II')-(V') ($T = 4.2$°K), shown as solid lines. The experimental points at 4.2°K, shown as open circles, will be seen to lie close to the theoretical curves. For the branch (V'), center of the total absorption curve is plotted instead of two observed peaks.

$J_0/k = 9.3$°K,[3] we find the theoretical curves shown by the solid lines in Fig. 2. It will be seen that they are in good coincidence with our experimental data.

This model of spin-cluster resonance is also supported by several other facts. First, the analysis predicts an exponential decrease in intensity with decreasing temperature at low temperatures, as is observed. This comes from the exponential decrease in n_e. Next, the observed intensity ratio of branches (III) and (V) near H_{c2} is about $\frac{1}{3}$, which is reasonably explained by the fact that the number of chains contributing to resonance branch (III) is $\frac{1}{3}$ of the number contributing to branch (V), as can be seen in Fig. 1(b). Finally, an anomalous behavior of (II) associated with the hysteresis of H_{c1} can be explained[5] satisfactorily although details are not given here. The angular dependence of the resonance shows that only the component of the field along the b axis is effective as is expected from an Ising spin model.

Several unsolved problems remain. The observed linewidths and shapes are somewhat different from those given by the theory, and frequently two peaks are observed on branch (V), as is shown in Fig. 1. The line splitting has not yet been explained. A detailed report will be published in the Journal of the Physical Society of Japan. The authors thank Professor Kanamori and Professor Tomita for useful discussions. They are also indebted to Professor E. Callen for reading the manuscript.

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[5] M. Motokawa and M. Date, J. Phys. Soc. Japan 20, 465 (1965). In this initial work, the authors found that two frequency-independent broad lines appeared near the critical fields. With the refined measurements discussed in this Letter, however, we show that these lines have slight frequency dependences, as are represented in Figs. 1 and 3.

[6] n_e is obtained in the following way: The total number $W(n_c, n_s)$ of independent arrangements of n_s spins in n_c clusters is ${}_{n_s-1}H_{n_c-1}$, where H is the homogeneous product $(n_s-1)!/(n_c-1)!\,(n_s-n_c)!$. Now let us

consider a case where n clusters are $m=1$ and the other (n_c-n) clusters of $m\geq 2$ have (n_s-n) spins as a whole. Then the number of independent states $w(n_c, n_s, n)$ is represented as $w={}_{n_c}C_n \times {}_{n_c-n}H_{n_s-n_c}$. Accordingly, the mean value of n, i.e., n_e, is given by

$$n_e=\sum_n nw/W=\sum_{n=1}^{nc-1} n\times {}_{n_c}C_n \times {}_{n_c-n}H_{n_s-n_c}.$$

Using a formula of combination products,

$$\sum_{r=0}^{q} {}_{\alpha}C_{q-r}\times {}_{\beta}C_r={}_{\alpha+\beta}C_q \quad (\alpha\geq q,\beta\geq q),$$

n_e reduces to $n_e=n_c(n_c-1)/(n_s-1)\simeq {n_c}^2/n_s$ when $n_c\gg 1$, $n_s\gg 1$. Similarly, the number of clusters having $m=\lambda$ is obtained as ${n_c}^2/n_s(1-n_c/n_s)^{\lambda-1}$.

[7]M. Date and M. Motokawa, Phys. Rev. Letters 15, 854 (1965).

SYSTEMATICS OF LOW-LYING 0^+ STATES IN LIGHT NUCLEI*

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Recently it has been suggested[1] that the 20.3-MeV 0^+ state of He^4 might be described by a "breathing mode." The present note calls attention to certain systematics[2] of 0^+ excited states in light even-even nuclei, particularly closed-shell nuclei, which show that a breathing-mode[3] description of these states is probably not successful. A more fruitful description in terms of multipair excitations, a pair consisting of a particle and a hole, is indicated.

Table I lists the 0^+ states considered here. Column 2 gives[4] excitation energies E. Column 3 presents a first systematic trend: The energies of these states follow roughly a $1/R^2$ dependence, where R is the nuclear radius. For a breathing mode one expects a $1/R$ energy dependence.[5] The argument can of course be made[1] that the He^4 state represents a breathing mode and that in the other nuclei the corresponding state, lying roughly at 20.3 $(4/A)^{1/3}$ MeV, has not yet been discovered.

Table I. 0^+–0^+ transitions in light nuclei. The energy of the transition is denoted by E, the pair width by Γ_π. The quantities ρ and f are the normalized monopole transition element and fraction of a monopole sum rule exhausted by the transition. Exact definitions are given in the text.

	E (MeV)	$EA^{2/3}$	Γ_π (eV)	ρ[e]	f (%)
He^4	20.3	51	$(3.4\pm0.9)\times10^{-4}$ [a]	0.23	11
C^{12}	7.66	40	$(6.2\pm0.6)\times10^{-5}$ [b]	0.52	16
O^{16}	6.05	38	$(9.2\pm0.9)\times10^{-6}$ [c]	0.31	4.0
Ca^{40}	3.35	39	$(1.9\pm0.1)\times10^{-7}$ [d]	0.12	0.25
Zr^{90}	1.75	35	$(2.2\pm0.2)\times10^{-9}$ [d]	0.048	...

[a] Ref. 6.
[b] Ref. 7.
[c] Ref. 8.
[d] Ref. 4.
[e] The percentage uncertainty in ρ is approximately one-half of that in Γ_π.

Column 4 in Table I lists the internal pair widths Γ_π of the states, taken from various references.[4,6-8] A second systematic trend can be noted by extracting from Γ_π the normalized transition matrix element $\rho=\langle r^2\rangle_{if}/R^2$, where $\langle r^2\rangle_{if}$ is the monopole matrix element between the initial state i and the final state f and R is the nuclear radius. As has been customary[9] we have put $R=(\frac{1}{2}e^2/mc^2)A^{1/3}=1.41A^{1/3}$ F. Using interpolations of calculations by Zirianova and Krutov,[10] the values for ρ in column 5 of Table I are found. One notes that the normalized transition matrix element for He^4 is comparable to that for C^{12}, O^{16}, and Ca^{40}. A similar conclusion can be drawn by substituting the experimental monopole matrix element into Ferrell's sum rule[11] for $T=0$ to $T=0$ monopole transitions:

$$N=\sum_f 2mE\,|\langle r^2\rangle_{if}|^2/(\hbar^2\langle r^2\rangle), \qquad (1)$$

where N = number of nucleons, m = mass of nucleon, $\langle r^2\rangle$ = mean square radius of ground state. One then finds that each of the 0^+-0^+ transitions considered here exhausts only a fraction f of the sum rule, shown in column 6 of Table I. [The fraction f is defined as the ratio of the right-hand side of Eq. (1) to the left-hand side.] It can be seen that the He^4 transition does not exhaust a greater portion of the sum rule than the C^{12} transition, and neither exhausts the full sum rule. Since the model discussed in Ref. 1 does exhaust[1] the full sum rule (1), it does not appear that the He^4 state is well de-