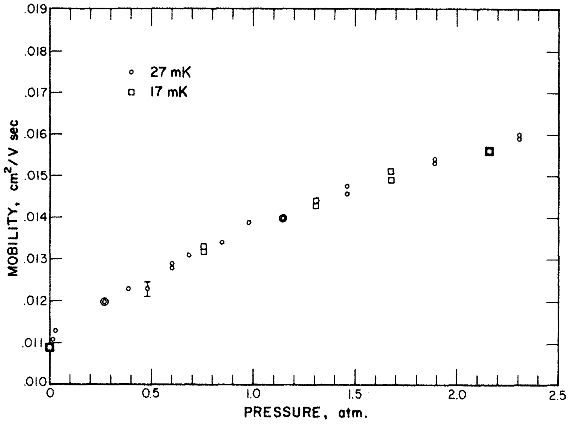

FIG. 2. Pressure dependence of the negative-ion mobility in pure ^{3}He at low temperature.

The low temperatures used in these experiments were produced by a ^{3}He-^{4}He dilution refrigerator with a copper mixing chamber. The mobility was measured with a double-gate velocity spectrometer with a 2.1-cm path length which was housed in a copper cell containing ~2000 cm^2 of sintered Cu for heat contact. Ions were produced by a tritiated titanium source of 3.5-mCi nominal intensity which introduced a heat input of 1.2 erg/sec. The temperature differential between the liquid and the chamber (calculated from the known Kapitza resistance) should thus be of the order of our temperature scale error. The temperature of the mixing chamber-ion cell combination was measured by a cerium-magnesium-nitrate magnetic thermometer made up of single crystals and coil foil in a roughly spherical shape. The magnetic thermometer was calibrated against the vapor pressure of ^{3}He between 0.6 and 1.5 K, and the error in the temperature scale is estimated to be 5%.

[†] Based on work performed under the auspices of the U.S. Atomic Energy Commission.

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Multiple-Quantum Transitions in a Rotating Magnetic Field*

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The multiple-quantum and rotating-field descriptions of resonant transitions requiring two frequencies are compared. Transitions combining features of both descriptions have been observed.

I. INTRODUCTION

Happer[1] has discussed two-frequency resonant transitions in which one may consider the field of one applied frequency ν_1 as establishing the stationary states and the field of the other frequency ν_2 as causing transitions between them. This description appears to differ somewhat from the idea of multiple-quantum transitions,[2] in which a transition between stationary states (established in the absence of applied frequencies) is produced by the absorption of several photons. In fact, Happer's treatment assumes that photons of frequency ν_1 are plentiful and those of frequency ν_2 are rare. All the transition frequencies he predicts can be written $\nu_2 = |(\Delta E_{ij}/h) + n\nu_1|$, where ΔE_{ij} is the energy difference between two levels and n is an integer; clearly such transitions involve one photon at ν_2 and n photons at ν_1. We have studied two-frequency transitions of types not previously observed, including types which require several photons of frequency ν_2.

II. THEORY

In this work the resonant system is an atom in a constant applied magnetic field H_c, which defines the z direction. Transitions between hyperfine levels are induced by two oscillating magnetic fields H_1 and H_2, both in the x direction. If the atom has electronic angular momentum $J=\frac{1}{2}$, the hyperfine Hamiltonian will be[3]

$$\mathcal{H}=hA(I\cdot J)+g_J\mu_0 J_z H_c +g_J\,\mu_0 J_x H_1\cos\omega_1 t+g_J\mu_0 J_x H_2\cos\omega_2 t\,. \quad (1)$$

The perturbation theory of multiple-quantum transitions[2,4] may be illustrated by calculating the transition probability from state k to state i through intermediate state j (Fig. 1). We expand the actual wave function of the atom in terms of ψ_n, the eigenstates of the first two terms of Eq. (1), and consider the last two terms as perturbations. Then we solve the time-dependent Schrödinger equation for the coefficient of ψ_j

$$\frac{da_j}{dt}=\frac{-i}{\hbar}a_k e^{(i/\hbar)(E_j-E_k)t}<\psi_j|\mathcal{H}|\psi_k>,$$

$$a_j=\frac{-1}{2\hbar}a_k<\psi_j|J_x|\psi_k>g_J\,\mu_0 H_1\frac{e^{i(\omega_{jk}-\omega_1)t}}{(\omega_{jk}-\omega_1)}$$

+ rapidly oscillating terms,

where $\omega_{jk}=(E_j-E_k)/\hbar$ is assumed to be almost equal to ω_1. Then the coefficient of ψ_i can be found:

$$\frac{da_i}{dt}=\frac{i}{4\hbar^2}a_k\langle\psi_i|J_x|\psi_j\rangle\langle\psi_j|J_x|\psi_k\rangle \times g_J^2\mu_0^2 H_1H_2\frac{e^{i(\omega_{ij}+\omega_{jk}-\omega_1-\omega_2)t}}{(\omega_{jk}-\omega_1)}+\cdots\,. \quad (2)$$

Resonance occurs when $\omega_{ij}+\omega_{jk}=\omega_1+\omega_2$, that is, when energy is conserved for the transition from k to i. Figure 1 is correct in displaying this energy conservation, but it is misleading to the extent that eigenstates k and i are affected by the oscillating fields H_1 and H_2. If these fields are strong enough to cause transitions, they will usually also cause energy-level shifts and wave-function admixtures which affect the observed resonances.[4] In the multiple-quantum picture all such effects must be computed separately by perturbation theory.

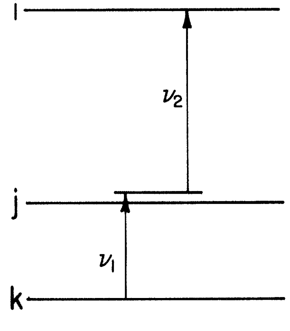

FIG. 1. Two-quantum transition from k to i.

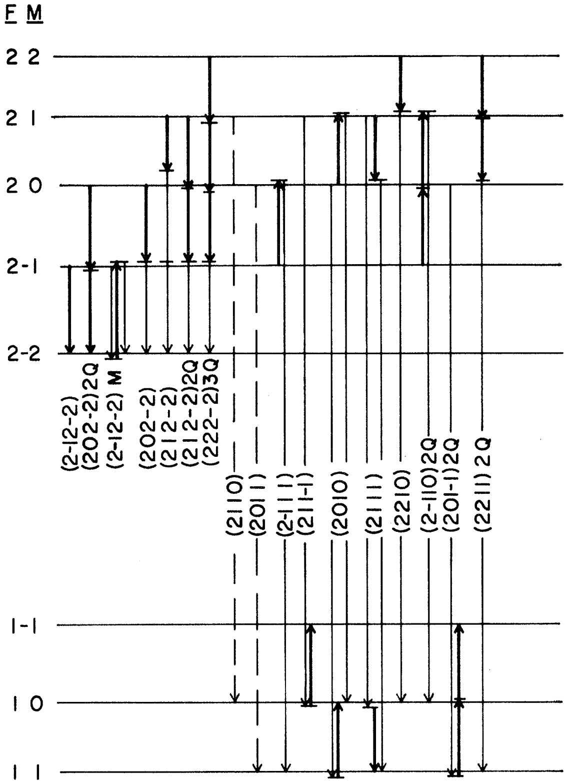

FIG. 2. Transitions observed in this work. Light arrows are ν_1 quanta and heavy arrows ν_2 quanta. The transitions are labeled by the quantum numbers of the initial and final states ($F_1M_1F_2M_2$) and the number of ν_2 quanta required. M is the mirror transition (see Ref. 1); the dashed lines do not involve ν_2 and are included for reference only.

Happer's analysis[1] transforms to a coordinate system rotating[5] at frequency ω_1 about the z axis. Then the first three terms of (1) can be used to compute eigenstates, and only H_2 (and the counter-rotating part of H_1) need be neglected or treated as perturbation. This procedure accounts quite accurately for the effect of H_1 on the energy levels and wave functions, and we have used it in the computer calculations which we compared with experiment. Happer[1] has obtained excellent agreement between similar calculations and the observed dependence of ν_2 on H_1 and ν_1, which indicates that the perturbations due to H_2 may often be neglected. In such a case the rotating-field description is clearly superior.

Many groups have observed transitions which take place by the absorption of more than one photon at a single frequency.[2] We report here the observation of such transitions (at ν_2) in the presence of another stronger field H_1 which perturbs the energy levels. Thus, the multiple-quantum resonant frequency ν_2 can most conveniently be calculated in the coordinate system rotating with H_1. However, we have chosen to represent the transitions on a diagram in the nonrotating system because it shows the photons at both ν_1 and ν_2. Figure 2 is such a diagram for the transitions observed in this experiment.

We remark in passing that the rotating-field and multiple-quantum pictures have also been developed for nuclear magnetic resonance.[6]

III. EXPERIMENT

The experiments were done with the atomic-beam apparatus described by Bernstein *et al.*[7] The rf loop which produced the transition was identical in design to Happer's. The main loop (H_1) was a circular solenoid about 2 cm long made of 12 turns of heavy wire, and the auxiliary loop (H_2) was a few turns of smaller wire wound around its center. Relative rf amplitudes were read from a voltmeter on the oscillator panel; the frequencies were measured with a cycle counter. The isotope used was K^{39}, and the magnetic field H_c was held at 13.706 G, corresponding to a frequency of 10.225 Mc/sec for the (2-12-2) transition (see Fig. 2).

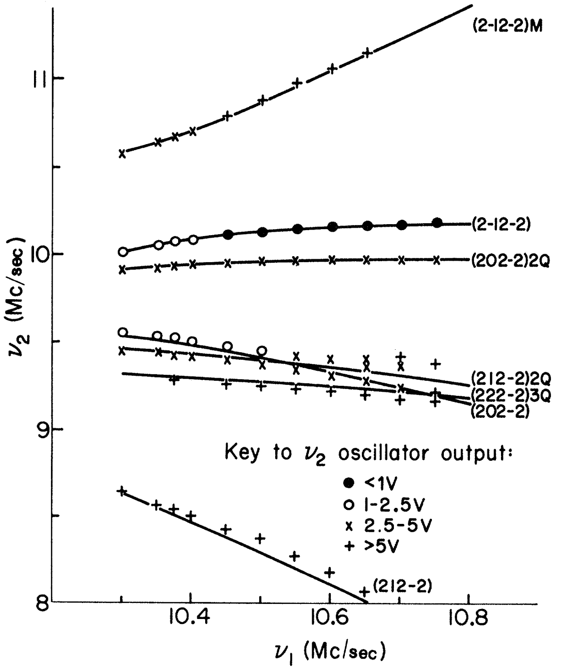

FIG. 3. Calculated and observed resonances near 10.5 Mc/sec.

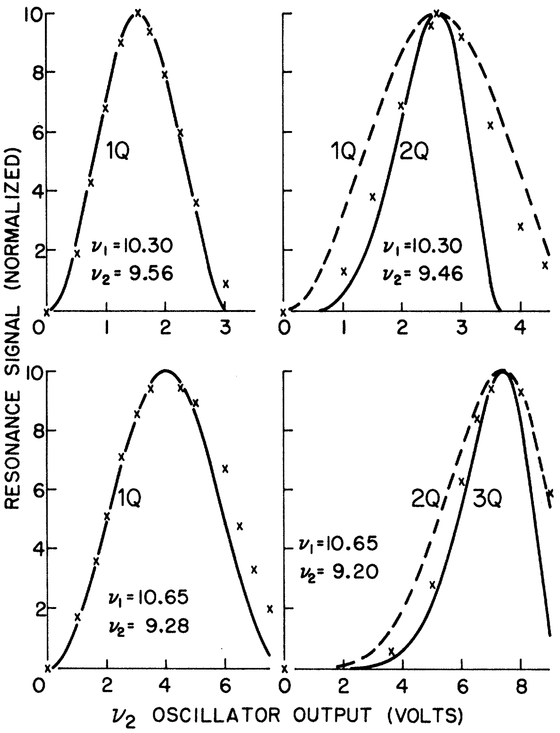

FIG. 4. Results of varying ν_2 oscillator output. The curves are normalized to the observed peak positions (see text).

Figure 3 shows the results of the first experiment. The points represent observed resonances ν_2 for various values of ν_1, and the lines show the calculated resonant frequencies. The ν_1 oscillator output was held at 3 V; the calculation uses $H_1 = 0.2$ G. The conversion factor 15 V/G came from a study of the variation of ν_2 with ν_1 voltage[1] at $\nu_1 = 10.3$ Mc/sec.

The most interesting part of Fig. 3 is the region where three resonances cross. Two of the three involve more than one quantum at frequency ν_2. The points do not lie exactly on the lines because both H_1 and H_2 perturb the energy levels, and the calculation only considers H_1. Near $\nu_2 = 9.38$ Mc/sec, the (2 1 2 0) resonance frequency, the perturbation due to H_2 is quite large; the same resonance was even found at different frequencies for different values of H_2. To confirm our assignment of the points to the lines, we tested their quantum multiplicity for ν_2 by measuring the dependence of transition probability on rf amplitude.[8] Figure 4 shows this dependence for selected points. Theoretically,[2] it should vary as $\sin^2(aH_2^N)$, where N is the multiplicity, but systematic devia-

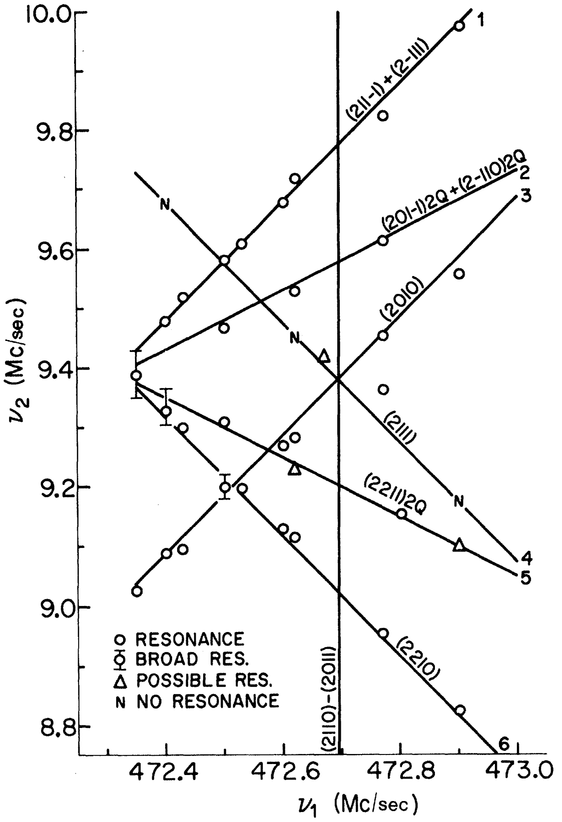

FIG. 5. Calculated and observed resonances near 472.7 Mc/sec.

tions can be expected at large H_2.[9] The results for small H_2 appear more consistent with the calculated multiplicities (solid curves) than with possible alternates (dashed curves).

The third multiple-quantum transition in Fig. 3 is the (2 0 2-2)2Q line. It can be observed in the absence of ν_1, and then has the frequency 9.999 Mc/sec. The presence of ν_1 can be seen to lower this frequency by the amount calculated.

For the second experiment, ν_1 was set near 472.7 Mc/sec, the resonant frequency of the transitions (2 1 1 0) and (2 0 1 1). The impedance of the 12-turn loop was quite large at this frequency, but a broad resonance was found at 5 V oscillator output. This may be compared with the 0.2 V needed at 10.225 Mc/sec. The oscillator output was increased to 22 V, and the two-frequency resonances shown in Fig. 5 were found. In this case, the lines represent the calculated frequencies at zero H_1.

Figure 5 contains a variety of interesting effects. Lines 2 and 5 are two-quantum transitions, easily identified by the different slopes. Lines 1 and 2 represent frequencies that can cause transitions by two mechanisms connecting different pairs of states; the transition intensities therefore add, and the observed resonances were somewhat larger than usual. Lines 3 and 4 represent frequencies that can cause transitions by two mechanisms connecting the same pair of states (see Fig. 2); the transition amplitudes therefore add. For line 4, the two transition amplitudes, computed to lowest order using Eq. (2), are equal and opposite, so that line 4 is forbidden. Although we searched carefully for it, it was observed only when ν_1 was very close to 472.7 Mc/sec. Here higher-order terms are important.

Transitions similar to these have been reported,[10] but with the roles of ν_1 and ν_2 reversed; the low-frequency signal perturbed the energy levels and the high-frequency resonance was split by the perturbation. This is sometimes called the Autler-Townes effect.[11]

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