SOVIET PHYSICS JETP VOLUME 18, NUMBER 4 APRIL, 1964

FREQUENCY CONVERSION IN QUANTUM SYSTEMS WITH DISCRETE ENERGY LEVELS

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Submitted to JETP editor May 21, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **45**, 1399-1407 (November, 1963)

An analysis, based on the equation of motion for the density matrix, is made of the various ways of performing frequency addition and multiplication using two and three discrete energy levels. Expressions are obtained for the efficiency of frequency conversion when the specimen is in a cavity tuned to the transformed frequency, and for the probabilities of the corresponding multiple-quantum transitions. Experiments consisting of frequency doubling in the 3-cm region using the Zeeman levels of ruby and of the free radical diphenylpicrylhydrazyl have confirmed the main conclusions of the theory.

INTRODUCTION

A number of works have recently appeared devoted to the study of parametric frequency changing with the aid of quantum systems with sharp spectral lines (see, for example, [1-6]). In the work of Bunkin[1] and of Fontana et al[2] parametric amplification and generation in two-level quantum systems[1] are analyzed; Faĭn et al[3] have considered subtraction of two frequencies in a three-level system. Fontana et al have studied both theoretically[4] and experimentally[5] frequency tripling in a two-level system where the frequency to be multiplied ω is approximately $1/3$ of the Bohr frequency of the system ω_0; the analysis was based on the Schrödinger wave equation. The case of frequency multiplication in two-level systems with $\omega \sim \omega_0$ is in essence used in ferromagnetic frequency multipliers (see, for example, [8]), where the analysis is based on equations of the Landau-Lifshitz type. The possibility of frequency doubling using three levels was experimentally observed by Kellington.[6]

Frequency conversion by quantum systems with sharp spectral lines is closely related to multiple-quantum transitions. Two-quantum induced transitions, frequently observed in radiospectroscopy (see, for example, [9]), show up as resonance absorption when the condition $|\omega_1 + \omega_2| \approx \omega_{21}$ is satisfied, where ω_1 and ω_2 are the frequencies of the two electromagnetic fields, $\omega_{21} = \hbar^{-1}(E_2 - E_1)$, and E_n is the level energy ($E_2 > E_1$). Such transitions can be divided into two types: in the first there is no real additional level with energy E_3 such that $|\omega_{31}| \approx \omega_1$ (or $|\omega_{31}| \approx \omega_2$); in the second type there is such a level. To these two types of multiple-quantum transitions correspond the possibilities of frequency conversion using two or three energy levels. It will be shown below [see (5) and (24)] that when $\omega_1 = \omega_2 \approx \omega_{21}/2$ the probability of transitions of the second type is, other conditions being equal, $(\tau\omega)^2$ times greater than the probability of transitions of the first type (τ is the relaxation time determining the line width).

Induced multiple-quantum transitions and various ways of performing frequency addition and multiplication are analyzed in the present paper. In the first section the theoretical method is given. In the second and third sections the theoretical results obtained for the cases of two and three levels are presented. In the fourth section the various ways of performing frequency multiplication are compared. Experimental results are given in the fifth section.

1. CALCULATION PROCEDURE

To be definite, the case of magnetic dipole interaction for a substance in a cavity is considered. The calculation is based on the simultaneous solution of the equation for the density matrix ρ, which allows the magnetization to be calculated from the formula $\mathbf{M} = N_0 \mathrm{Sp}(\mu\rho)$, and the equation for the cavity oscillations; this gives for steady-state conditions the relation between the complex amplitudes of the magnetization vector $\mathbf{M}_k$ and the field $\mathbf{H}_k$:

$$\mathbf{H}_k = -i4\pi Q_L \eta \mathbf{h}_k (\mathbf{h}_k \mathbf{M}_k);$$

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[1] Parametric effects in a two-level system are in essence also used in ferromagnetic amplifiers and generators when there is homogeneous precession of the magnetization.[7]

Here N_0 is the number of particles per unit volume of the substance, μ is the magnetic moment operator for the particle, Q_L is the quality factor of the loaded cavity, η is the filling factor, $\mathbf{h}_k$ is a unit vector in the direction of the magnetic field for the oscillation mode of frequency ω_k in the cavity region occupied by the substance (it is assumed that the field is homogeneous in the specimen and that one of the characteristic frequencies of the cavity is equal to the corresponding frequency of oscillation of **M**; this relation is valid under the assumption that the perturbation of the characteristic frequencies of the cavity by the substance is small).

The equation for the density matrix includes phenomenological relaxation terms (see, for example, [1,3,10-13]) characterized by the spin-spin relaxation times τ_{nm} and by the probabilities of transitions due to interaction with the thermostat w_{nm} and interaction with the external fields expressed in the form

$\frac{1}{2}\sum [\mathbf{H}_k e^{i\omega_k t}$ + complex conjugate λ], so that the energy of interaction is

$$\hbar \sum [A^{(k)} e^{i\omega_k t} + A^{(k)+} e^{-i\omega_k t}],$$

where $A^{(k)} = -\mu \mathbf{H}_k / 2\hbar$.

The values of $\mathbf{H}_k$ and $\mathbf{M}_k$ obtained allow the power at frequency ω_k emitted (or absorbed, depending on the sign) by the substance to be calculated from the formula $P_k = \frac{1}{2} V \,\mathrm{Re}\,(i\omega_k \mathbf{M}_k \cdot \mathbf{H}_k^*)$ where V is the specimen volume. The situation $P_k < 0$ corresponds to emission; in order to obtain the useful power in the exit wave guide, it is necessary to multiply P_k by Q_L/Q_e, where Q_e is the quality factor of the coupling.

2. TWO-LEVEL SYSTEM

Let the external field contain frequencies ω_1 and ω_2 where $\omega_1 + \omega_2 \equiv \omega_3 \sim \omega_0$ ($\hbar\omega_0 = E_2 - E_1$). A steady state solution of the equation for the density matrix is sought in the form

$$\rho_{12} = \sum_n \rho_n e^{i\omega_n t}, \quad n = \pm 1, \pm 2, + 3; \quad \omega_{-n} = -\omega_n;$$

$$\Delta = \rho_{11} - \rho_{22} = \Delta^{(0)} + \sum_k \Delta_k e^{i\omega_k t},$$

$$k = \pm 1, \pm 2; \quad \Delta_{-k} = \Delta_k^*.$$

The theoretical results are conveniently presented in the following form:

$$P_1 = \Delta^{(0)} N\hbar\omega_1 (W_1' - W_2'' + W), \tag{1}$$

$$P_2 = \Delta^{(0)} N\hbar\omega_2 (W_2' - W_2'' + W), \tag{2}$$

$$P_3 = -\Delta^{(0)} N\hbar\omega_3 W Q_L/(Q_L + Q_m), \tag{3}$$

$$W_k' = \frac{2\tau |A_{12}^{(k)}|^2}{1 + \tau^2 (\omega_k - \omega_0)^2}, \quad W_k'' = \frac{2\tau |A_{21}^{(k)}|^2}{1 + \tau^2 (\omega_k + \omega_0)^2}, \quad k = 1, 2; \tag{4}$$

$$W = \frac{2\tau' |A_{12}^{(2)} (A_{11}^{(1)} - A_{22}^{(1)}) / \omega_1 + A_{12}^{(1)} (A_{11}^{(2)} - A_{22}^{(2)}) / \omega_2 |^2}{1 + \tau'^2 (\omega_3 - \omega_0 - \delta_1)^2}; \tag{5}$$

$$\delta_1 = (2 |A_{12}^{(1)}|^2 - |A_{11}^{(2)} - A_{22}^{(2)}|^2) / \omega_2$$
$$+ (2 |A_{12}^{(2)}|^2 - |A_{11}^{(1)} - A_{22}^{(1)}|) / \omega_1; \tag{6}$$

$$\tau' = \tau Q_m/(Q_L + Q_m), \tag{7}$$

N is the total number of particles, $Q_m = (4\pi\hbar^{-1}\eta\tau\Delta^{(0)} N_0 |\mathbf{h}_3 \cdot \mu_{12}|^2)^{-1}$ is the magnetic quality factor. It is assumed that ω_1 and $\omega_2 \gg \tau^{-1}$ and $|A_{nm}^{(k)}| \ll \omega_k$. The quantity δ_1 is a correction to the resonance frequency analogous to the Bloch-Siegert shift.[14] The factor $Q_m/(Q_L + Q_m)$ takes into account radiative damping.[15]

The steady-state part of the equation for the diagonal elements of the density matrix reduces to the form

$$d\rho_{11}^{(0)}/dt = \{w_{21} + W_1' + W_1'' + W_2' + W_2''$$
$$+ W [1 - Q_L/(Q_L + Q_m)]\} \rho_{22}^{(0)} - \{w_{12} + W_1' + W_1''$$
$$+ W_2' + W_2'' + W [1 - Q_L/(Q_L + Q_m)]\} \rho_{11}^{(0)} = 0. \tag{8}$$

From (1), (2), (3), and (8) it follows that W_1' and W_2' can be interpreted as the probabilities of a transition from level 1 to level 2 with the absorption of quanta $\hbar\omega_1$ and $\hbar\omega_2$ (or as the probabilities of the inverse process); W_1'' and W_2'' are the probabilities of transitions 1 → 2 with emission of quanta; W is the probability of a two-quantum transition 1 → 2 (or 2 → 1) with simultaneous absorption (emission) of quanta $\hbar\omega_1$ and $\hbar\omega_2$. According to (3) the probability of emitting a quantum $\hbar\omega_3$ is proportional to the probability of two-quantum absorption W.

In what follows we consider the case when the external field has a frequency $\omega \sim \omega_0/2$. This variant is a particular case of the preceding. The power absorbed is

$$P_\omega = \Delta^{(0)} N\hbar\omega (W' - W'' + 2W), \tag{9}$$

where for W' and W'' we can use equations (4), in which we must omit the index k. The radiated power is

$$P_{2\omega} = \Delta^{(0)} N 2\hbar\omega W Q_L/(Q_L + Q_m), \tag{10}$$

$$W = \frac{2\tau' |A_{12} (A_{11} - A_{22})|^2 / \omega^2}{1 + \tau'^2 (2\omega - \omega_0 - \delta_2)^2}, \tag{11}$$

$$\delta_2 = (2|A_{12}|^2 - |A_{11} - A_{22}|^2)/\omega. \qquad (12)$$

The equations for population balance have the form

$$d\rho_{11}^{(0)}/dt = \{w_{21} + W' + W'' + W[1 - Q_L/(Q_L + Q_m)]\}\,\rho_{22}^{(0)}$$
$$- \{w_{12} + W' + W''$$
$$+ W[1 - Q_L/(Q_L + Q_m)]\}\,\rho_{11}^{(0)} = 0. \qquad (13)$$

It follows from (10) and (11) that, if the particles do not possess a constant dipole moment, frequency doubling is impossible (in this case multiple-quantum transitions of even order are in general impossible). The calculation of frequency tripling when $\omega \sim \omega_0/3$ reduces to equations (9) and (10), in which the factor 2 must be replaced by 3, and for W must be understood the probability of a three-quantum transition:

$$W = \frac{\tau' |A_{12}^2 A_{21}|^2 / 8\omega^4}{1 + \tau'^2(3\omega - \omega_0 - \delta_3)^2}, \qquad (14)$$

$$\delta_3 = (|A_{21}|^2 + 2|A_{12}|^2)/2\omega. \qquad (15)$$

The population balance equations are of the form of (13) as previously. The expression obtained from (10), (11) and (12) for the output power of frequency 3ω when the fields $\mathbf{H}_\omega$ and $\mathbf{H}_{3\omega}$ are linearly polarized and parallel to μ_{12}, and when it can be considered that $\tau' = \tau$, agrees with the corresponding formula in [4].

We give formulae for the harmonic emission powers when $\omega \sim \omega_0$. The second harmonic emission power is

$$P_{2\omega} = 4\pi\omega \frac{Q_L f}{V_{\text{cav}}} (\Delta^{(0)} N)^2$$
$$\times \frac{\tau^2 |A_{12}|^2 |\mathbf{h}_{2\omega}[(\mu_{11} - \mu_{22}) A_{21} - 2\mu_{21}(A_{11} - A_{22})]|^2}{\omega^2[1 + \tau^2(\omega - \omega_0 - \delta)^2]}, \qquad (16)$$

$$\delta = (|A_{21}|^2 + |A_{11} - A_{22}|^2)/\omega, \qquad (17)$$

where $f = \eta V_{\text{cav}}/V$ is a coefficient of order 2. We note that the expression for δ when $\mathbf{H}_\omega$ is linearly polarized and when $A_{11} - A_{22} = 0$ coincides with the value of the resonance shift obtained by Bloch and Seigert.[14]

If the diagonal elements of μ are zero the third harmonic emission power is

$$P_{3\omega} = 6\pi\omega \frac{Q_L f}{V_{\text{cav}}} (\Delta^{(0)} N)^2 \frac{\tau^2 |A_{12}^2 A_{21}|^2 |\mathbf{h}_{3\omega} \mu_{21}|^2 / \omega^4}{1 + \tau^2(\omega - \omega_0 - \delta)^2}. \qquad (18)$$

In the general case when $\omega \sim \omega_0$ the emitted power for the harmonic $n\omega$ is of order

$$P_{n\omega} \sim (Q_L/Q_m)\,|A_{ik}/\omega|^{2(n-1)} P_\omega, \qquad (19)$$

where P_ω is the power absorbed.

3. THREE-LEVEL SYSTEM

We consider a three-level system with energies $E_1 < E_2 < E_3$, on which acts an external field with frequencies $\omega_1 \sim \omega_{21}$ and $\omega_2 \sim \omega_{32}$ ($\hbar\omega_{nm} = E_n - E_m$). A steady state solution of the equation for the density matrix is sought in the form

$$\rho_{12} = \lambda_{12} e^{i\omega_1 t}, \quad \rho_{23} = \lambda_{23} e^{i\omega_2 t}, \quad \rho_{13} = \lambda_{13} e^{i\omega_3 t},$$
$$\rho_{nn} = \lambda_n; \quad \lambda_n - \lambda_m = \Delta_{nm}, \quad \omega_3 = \omega_1 + \omega_2.$$

Calculation leads to the following result:

$$P_{\omega_1} = N\hbar\omega_1(\Delta_{12} W_{12} + \Delta_{13} W_{13}), \qquad (20)$$
$$P_{\omega_2} = N\hbar\omega_2(\Delta_{23} W_{23} + \Delta_{13} W_{13}), \qquad (21)$$
$$P_{\omega_3} = -8\pi\omega_3\, Q_L f V_{\text{cav}}^{-1} N^2 |\mu_{13}\mathbf{h}_{3\omega}|^2 |\lambda_{13}|^2, \qquad (22)$$

where

$$W_{12} = 2\tau_{12} |A_{12}^{(1)}|^2/(1 + \sigma_{21}^2) - 2\tau_{13}' \tau_{12} |A_{12}^{(1)} A_{23}^{(2)}|^2$$
$$\times \operatorname{Re}\{[\tau_{12}/(1 + i\sigma_{21})$$
$$+ \tau_{23}/(1 + i\sigma_{32})]/(\xi + i\varphi)(1 + i\sigma_{21})\}, \qquad (23)$$
$$W_{23} = 2\tau_{23} |A_{23}^{(2)}|^2/(1 + \sigma_{32}^2)$$
$$-2\tau_{13}' \tau_{23} |A_{12}^{(1)} A_{23}^{(2)}|^2 \operatorname{Re}\{[\tau_{23}/(1 + i\sigma_{32})$$
$$+ \tau_{12}/(1 + i\sigma_{21})]/(\xi + i\varphi)(1 + i\sigma_{32})\},$$
$$W_{13} = 2\tau_{12}\tau_{23}\tau_{13}' |A_{12}^{(1)} A_{23}^{(2)}|^2$$
$$\times \operatorname{Re}\{(1 + i\sigma_{21})(1 + i\sigma_{32})(\xi + i\varphi)\}^{-1}, \qquad (24)$$

$$\lambda_{13} = \tau_{13}' A_{12}^{(1)} A_{23}^{(2)} [\tau_{23}\Delta_{23}/(1 + i\sigma_{32})$$
$$- \tau_{12}\Delta_{12}/(1 + i\sigma_{21})]/(\xi + i\varphi),$$
$$\xi = 1 + \tau_{13}' \tau_{23} |A_{12}^{(1)}|^2/(1 + \sigma_{32}^2) + \tau_{13}' \tau_{12} |A_{23}^{(2)}|^2/(1 + \sigma_{21}^2),$$
$$\varphi = \sigma_{31} - \sigma_{32}\tau_{13}' \tau_{23} |A_{12}^{(1)}|^2/(1 + \sigma_{32}^2)$$
$$- \sigma_{21}\tau_{13}' \tau_{12} |A_{23}^{(2)}|^2/(1 + \sigma_{21}^2),$$
$$\times \quad \sigma_{21} = \tau_{12}(\omega_1 - \omega_{21}), \quad \sigma_{32} = \tau_{23}(\omega_2 - \omega_{32}),$$
$$\sigma_{31} = \tau_{13}'(\omega_3 - \omega_{31}),$$
$$\tau' = \tau Q_m/(Q_L + Q_m), \quad Q_m^{-1} = 4\pi\hbar^{-1}\eta\tau_{13}\Delta_{13}N_0 |\mu_{13}\mathbf{h}_{3\omega}|^2. \qquad (25)$$

From the equations for the diagonal elements of the density matrix we obtain the following relations for the population balances of the levels:

$$d\lambda_1/dt = (w_{21} + W_{12})\lambda_2 + (w_{31} + W_{13})\lambda_3$$
$$- (w_{12} + W_{12} + w_{13} + W_{13})\lambda_1 - P_{\omega_3}/N\hbar\omega_3 = 0,$$
$$d\lambda_2/dt = (w_{12} + W_{12})\lambda_1 + (w_{32} + W_{23})\lambda_3 - (w_{21} + W_{12}$$
$$+ w_{23} + W_{23})\lambda_2 = 0,$$
$$\lambda_1 + \lambda_2 + \lambda_3 = 1. \qquad (26)$$

It follows from (20), (21), and (26) that W_{12} and W_{23} can be regarded as the probabilities of transitions 1 → 2 and 2 → 3, respectively, with the absorption of a single quantum (or the reverse transitions with emission), and W_{13} as the probability of the transition accompanied by a two-quantum process. Transitions 3 → 1 with the emission of a quantum $\hbar\omega_3$ also affect the population balances.

It is possible to obtain, from the formulae given, expressions for the case when an external field with a single frequency ω acts on the system, where $\omega \sim \omega_{21} \sim \omega_{32} \sim \omega_{31}/2$; to do this it is sufficient to put $\omega_1 = \omega_2 = \omega$ and $A^{(1)} = A^{(2)} = A$.

4. COMPARISON OF THE EFFICIENCY OF FREQUENCY MULTIPLICATION FOR THE DIFFERENT METHODS CONSIDERED

The formulae obtained for the emission power for arbitrary field polarizations and dipole moments are quite cumbersome. For the assessments we give expressions for the amplitude of the magnetization vector **M** for the various ways of frequency doubling and tripling; the formulae were obtained under the simplifying assumptions given below in brackets; in particular it is assumed that the ratios of the frequencies of the system and of the external field are optimum and that the external field is linearly polarized.

The relation between $\mathbf{M}_{n\omega}$ and the output power when the substance is in a cavity tuned to the frequency $n\omega$ is given by the formula

$$(P_{n\omega})_e = 2\pi n\omega V^2 |\mathbf{M}_{n\omega}\mathbf{h}_{n\omega}|^2\, Q_L f / Q_e V_{\text{cav}}. \tag{27}$$

1) $\omega = (\omega_0 + \delta)/n$:

$$|\mathbf{M}_{2\omega}| = \beta\Delta N_0\, (\beta H_\omega/\hbar)^2/\omega\, (\Delta\omega)' \tag{28}$$

($\mu = 2\beta\mathbf{S}$, $S = 1/2$); the field $\mathbf{H}_\omega$ makes an angle of 45° with the constant field **H**; $\mathbf{M}_{2\omega}$ rotates with frequency 2ω in the plane perpendicular to **H**; $(\Delta\omega)'$ is the width of the absorption line taking into account radiative damping [for a cavity $(\Delta\omega)' = \Delta\omega(Q_L + Q_m)/Q_m$],

$$|\mathbf{M}_{3\omega}| = \beta\Delta N_0\, (\beta H_\omega/\hbar)^3/8\omega^2\, (\Delta\omega)' \tag{29}$$

($\mu_{12} = \beta\mathbf{h}_\omega$, $\mu_{11} = \mu_{22} = 0$; $\mathbf{M}_{3\omega}$ oscillates parallel to $\mathbf{H}_\omega$);

2) $\omega = \omega_0 + \delta$:

$$|\mathbf{M}_{2\omega}| = \beta\Delta N_0\, (\beta H_\omega/\hbar)^2/\omega\Delta\omega \tag{30}$$

($\mu = 2\beta\mathbf{S}$, $S = 1/2$, $\mathbf{H}_\omega \perp \mathbf{H}$, $\mathbf{M}_{2\omega} \parallel \mathbf{H}$),

$$|\mathbf{M}_{3\omega}| = \beta\Delta N_0\, (\beta H_\omega/\hbar)^3/4\omega^2\Delta\omega \tag{31}$$

($\mu_{12} = \beta\mathbf{h}_\omega$, $\mu_{11} = \mu_{22} = 0$, $\mathbf{M}_{3\omega} \parallel \mathbf{H}_\omega$),

3) $\omega = \omega_{32}/2 = \omega_{21} \pm \Delta\omega/2 = \omega_{32} \mp \Delta\omega/2$:

$$|\mathbf{M}_{2\omega}| = 2\beta\Delta N_0\, (\beta H_\omega/\hbar)^2/(\Delta\omega)^2 \tag{32}$$

($\mu_{ij} = \beta\mathbf{h}_\omega$, $\tau_{ij} = \tau$, $\Delta_{21} = \Delta_{32} = \Delta$; we neglect radiation reactions; $\mathbf{M}_{2\omega} \parallel \mathbf{H}_\omega$).

It follows from the formulae given that, if we neglect corrections due to radiative damping, the effect of saturation, and the effect of absorption in the substance on the value of $\mathbf{H}_\omega$, then the efficiency of transformation is practically equal in cases 1) and 2) and (for frequency doubling) is $(\omega/\Delta\omega)^2$ times smaller than in case 3).

5. EXPERIMENT

Frequency doubling in the 3-cm range was experimentally studied using the Zeeman levels of ruby and of the free radical diphenylpicrylhydrazyl (DPPH) at room temperature. The specimens were placed in a rectangular cavity for which the frequencies of the H_{011} and H_{103} modes of oscillation were, respectively 9.5 and 19 Gc (the unloaded quality factors Q_0 were about 5000 and 3000, respectively).

The experiments with DPPH confirmed the possibility of the two regimes of frequency doubling: for $H = \omega/\gamma$ and for $H = 2\omega/\gamma$ (H is the strength of the constant magnetic field and γ is the gyromagnetic ratio for DPPH). The polycrystalline specimens of DPPH were placed in the part of the cavity in which $\mathbf{h}_\omega$ and $\mathbf{h}_{2\omega}$ were perpendicular; rotation of the magnet in the plane $\mathbf{h}_\omega$, $\mathbf{h}_{2\omega}$ made it possible to change the angle φ between **H** and $\mathbf{h}_\omega$.

In Fig. 1 are given in arbitrary units the functions $(P_{2\omega}(\varphi))^{1/2}$ for $H = \omega/\gamma$ (Fig. 1a, $(P_{2\omega})^{1/2} \sim \sin\varphi(1 + \cos^2\varphi)$) and for $H = 2\omega/\gamma$ (Fig. 1b, $(P_{2\omega})^{1/2} \sim \sin\varphi\cos^2\varphi$), calculated from formulae (16) and (10), respectively; in the calculation it was assumed that $\mu_{nm} = -2\beta S_{nm}$ (β is the Bohr

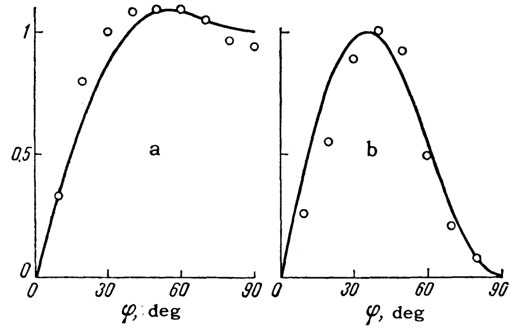

FIG. 1. Variation of $(P_{2\omega})^{1/2}$ (in arbitrary units) for DPPH with the angle ϕ between **H** and $\mathbf{H}_\omega$ ($\mathbf{H}_{2\omega} \perp \mathbf{H}$, $\mathbf{H}_{2\omega} \perp \mathbf{H}_\omega$) for $H = \omega/\gamma$(a) and $H = 2\omega/\gamma$ (b); continuous curve – theory; points – experiment.

magneton and S_i are the Pauli matrices); it was also assumed that there was no saturation and that $Q_0 \ll Q_m$. Experimental points are also plotted in Fig. 1. They agree satisfactorily with the theoretical curves.

The measured transformation efficiencies $P_{2\omega}/P_\omega^2$ for optimum values of φ with no saturation and with $Q_0 \ll Q_m$ also agree in order of magnitude with the result of calculations using (10) and (16); in both cases they were $\sim 10^{-5}p^2\ W^{-1}$, where p is the weight of the specimen in grams.

To verify formula (22) the experiment briefly described by Kellington[6] on frequency doubling using three Zeeman levels of ruby was repeated. A single crystal of ruby, with a volume of 0.24 cm^3 and with a Cr^{3+} ion concentration of 0.05% was used in the experiment.

In a constant magnetic field **H** the Cr^{3+} ions in ruby have four spin levels with energies E_n; the corresponding Bohr frequencies $\omega_{nm} = \hbar^{-1} \times (E_n - E_m)$ are functions of the value of H and of the angle θ between **H** and the crystal axis. From a consideration of the functions $\omega_{nm}(\theta)$ which are determined by the spin-Hamiltonian for ruby,[16] it can be shown that the condition $\omega_{kl} \approx \omega_{lm} \approx \omega$ $(k < l < m)$ is satisfied when ω is in the 3 cm region for four pairs of values of H and θ: 1.5 kOe, 33°; 2.1 kOe, 90°; 3.4 kOe, 65°, and 4.6 kOe, 28°.

The variation of the second harmonic power $P_{2\omega}$ with H and θ was measured in the experiment for a fixed frequency ω; it is, therefore, convenient to transform (22), taking H and θ as independent variables. In the neighborhood of the point $H' = 3.44$ kOe, $\theta' = 65°$, which was studied in detail, the function $\omega_{nm}(\theta)$ is nearly linear; let $\omega_{nm}(\theta) = \omega - \gamma_{nm}(\theta)[H_{nm}(\theta) - H]$, where $\gamma_{nm} = \partial\omega_{nm}/\partial H$, $H_{21}(\theta)$ and $H_{32}(\theta)$ are the resonance fields of the corresponding transitions for frequency ω, and $H_{31}(\theta)$ is the same quantity for frequency 2ω. In the neighborhood of the specified point for $\omega = 2\pi \times 9.5$ Gc the function $H_{nm}(\theta)$ approximately satisfies the relation $H_{21} - H_{31} = -(H_{32} - H_{31}) = k(\theta - \theta')$ $(k \approx 132$ Oe/deg$)$ and the coefficients γ_{nm} are almost independent of θ, where $\gamma_{21} \approx \gamma_{32} \approx \gamma_{31}/2 \equiv \gamma = 2\pi \times 2.28$ Gc/kOe. The experimental values of the absorption line widths at the specified point were $\Delta H_{21} = 44$ Oe, $\Delta H_{32} = 30$ Oe, and $\Delta H_{31} = 20$ Oe; if we take $\tau_{nm} = 2/\gamma_{nm}\Delta H_{nm}$, then the τ_{nm} are, respectively, 3.2, 4.7 and 3.5×10^{-9} sec.

To simplify the analysis we put $\tau = \Sigma\tau_{nm}/3 \approx 3.8 \times 10^{-9}$ sec. Then, taking into account that $|A_{nm}|\tau \ll 1$, $\Delta_{12} = \Delta_{23}$ and using the notation $x \equiv \gamma\tau(H_{31} - H)$, $a \equiv \gamma\tau(H_{31} - H_{21})$ we obtain from equation (22) when $\omega_1 = \omega_2 = \omega$ the following formula, which determines the variation of $P_{2\omega}$ with $H - H_{31}$ and $\theta - \theta'$:

$$f(x, a) = 4a^2/(1 + 4x^2)[1 + (x - a)^2][1 + (x + a)^2]. \tag{33}$$

The variation of f(x, a) with x for a = const has a principal maximum at x = 0 (i.e., when $\omega_{31} = 2\omega$), and, if $|a| \gtrsim 3.7$, has two additional maxima at $x = \pm a$ (i.e. when $\omega_{21} = \omega$ and $\omega_{32} = \omega$), where $f(0, a) \approx 16f(a, a) \approx 4/a^2$. The variation of f(x, a) with a when x = 0 has a double-peaked form with a minimum of zero at a = 0 and two maxima equal to 1 at $a = \pm 1$. Thus, $P_{2\omega}$ is a maximum, not when the levels are strictly equidistant and there is resonance $(\omega = \omega_{31}/2 = \omega_{21} = \omega_{32})$, but when $\omega = \omega_{31}/2 = \omega_{21} \pm \Delta\omega/2 = \omega_{32} \mp \Delta\omega/2$, where $\Delta\omega = 2/\tau$ is the absorption line width.

Experiment confirmed the features of the function $P_{2\omega}(H, \theta)$ considered above. For $\theta_{1,2} = \theta' \pm 0.17°$ and $H = H_{31}$ two almost equal maxima of $P_{2\omega}$ were observed, which agrees satisfactorily with the theoretical condition $a_{1.2} = \pm 1$ $(a \approx 7.3\,(65° - \theta°)$, so that $a_{1.2\text{exp.}} \approx \mp 1.2)$; $P_{2\omega}(H_{31}, \theta')$ was more than one order smaller than $P_{2\omega}(H_{31}, \theta' \pm 0.17°)$. Further, the emission line width $\Delta H_{2\omega}$ at the frequency 2ω when $a = \pm 1$ should, according to (33), be $\Delta x \approx 1$, i.e. $\Delta H_{2\omega} = 18$ Oe, and the experimental value was $\Delta H_{2\omega} = 15$ Oe.

In Fig. 2 is given a specimen oscilloscope trace showing the variation of $(P_{2\omega})^{1/2}$ with H when $\theta - \theta' = 0.5°$ $(a \approx 3.5)$; the principal emission peak corresponds to the condition $\omega = \omega_{31}/2$, and the two side maxima to $\omega = \omega_{21}$ and ω_{32}.

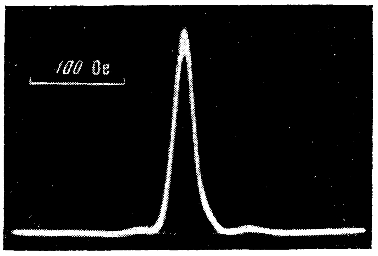

FIG. 2. Variation of $(P_{2\omega})^{1/2}$ with magnetic field for ruby. The principal maximum corresponds to the condition $\omega_{31} = 2\omega$, the two additional maxima to the conditions $\omega_{21} = \omega$ and $\omega_{32} = \omega$.

The experimental transformation efficiency $P_{2\omega}/P_\omega^2$ for optimum H and θ in the absence of saturation was $\sim 10^{-7}W^{-1}$; an estimate from formula (22) gave $P_{2\omega}/P_\omega^2 = 5 \times 10^{-7}W^{-1}$ (the values of μ_{nm} were determined from the spin-Hamiltonian).

CONCLUSION

The satisfactory agreement between experimental and theoretical results allows one to conclude that, despite the crude way in which the interactions of the particles with one another and with the thermostat were taken into account by using relaxation times, the method of analysis which was used allows one to evaluate the various ways of transforming frequency using a system of weakly interacting particles possessing a constant or induced dipole moment.

In the present work the possibility was demonstrated both theoretically and experimentally of three ways of frequency doubling: 1) the frequency to be multiplied close to a Bohr frequency of the system ω_{21}; 2) the multiplied frequency 2ω close to ω_{21}; and 3) $\omega \sim \omega_{21}$ and simultaneously 2ω close to another frequency of the system ω_{31}. From the viewpoint of such a classification frequency multiplication due to non-linear electronic polarization of dielectric crystals (see, for example, [17]) can belong to a fourth way, in which both the original and the combination frequencies are far from characteristic frequencies of the system (which are determined in the case of dielectrics by the width of the forbidden gap). An analogous classification can also be made for other types of parametric transformation, for example, for subtraction, amplification and generation (the subdivision of ferromagnetic generators and amplifiers into electromagnetic, semistatic and static (see, for example, [8]) has much in common with the proposed classification).

The choice of the optimum method in practical devices should be made taking into account the specific features of the problem; it should be remembered that in the three methods enumerated there is resonance absorption and, consequently, strong dispersion in the region of either the initial frequency (see 1 above) or the transformed frequency (2) or these and other frequencies (3); dispersion plays a determining role in the case of travelling wave transforming devices (see, for example, [17]).

The authors thank S. A. Akhmanov, and R. B. Khokhlov for proposing the subject and discussing the problem, and also V. S. Buklanov and A. N. Penin for help with preparing and carrying out the experiments.

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Translated by K. F. Hulme
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