FREQUENCY SUBTRACTION BY MEANS OF A THREE-LEVEL SYSTEM

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The resonance subtraction of two frequencies in a quantum system with three discrete energy levels has been investigated theoretically and experimentally. The effect is regarded as the result of a three-photon process; the quantum-mechanical treatment allows us to use a diagram technique. The experiment consists of observing a field at a frequency of 70 MHz which is induced in a ruby single crystal when it is irradiated by two fields at frequencies of 10.22 and 10.15 GHz; the experimental results confirm the basic predictions of the analysis.

1. INTRODUCTION

IT is known that the efficiency of nonlinear processes in the interaction of radiation with matter increases sharply when the frequencies of the effective fields approach some characteristic frequency of the system ω_{mn}. For example, if a medium which does not have a center of symmetry is subject to two monochromatic fields with frequencies ω_1 and ω_2, where $\omega_1 \approx \omega_{31}$ and $\omega_2 \approx \omega_{32}$, a strong polarization component (or magnetization component) arises at the frequency $\omega_1 - \omega_2 \approx \omega_{21}$. This effect can be used for frequency subtraction in the optical and microwave regions. A detailed analysis of this effect based on the use of the density matrix is given in [1-4].

In Sec. 2 of the present paper, the subtraction of two frequencies by means of a three-level system is treated from the quantum-mechanical point of view; the probability for three-photon processes is computed by means of a diagram technique. In Sec. 3 we give the results of an experimental investigation of resonance subtraction of two frequencies in the microwave region ($\lambda - 3$ cm) using spin levels in the ground state of ruby; the frequency difference is 17 MHz.

2. QUANTUM-MECHANICAL ANALYSIS

The formulas for frequency subtraction in a three-level system can be found from the usual density matrix formalism for addition[5] by using the following interchange of subscripts and frequency ω_2: $2 \to 3$, $3 \to 2$, $\omega_2 \to -\omega_2$. The addition scheme is converted into a subtraction scheme by interchanging levels 2 and 3. The corresponding expression for the power at the difference frequency as governed by assumptions appropriate for the present experiment is given in the next section.

We shall consider in greater detail another method which is based on transition probabilities and field quantization. This method yields the possibility of unique interpretation of multiphoton processes; moreover, in conjunction with a diagram technique is very convenient for computing processes in higher approximations of perturbation theory (cf. for example [6], where quantum-mechanical absorption in a two-level system is treated in detail). Certain rules for forming the matrix element through the graph technique are given in [7]. In [6] and [7] consideration is given to processes by which a particle makes a transition to a different level. We can introduce the notion of a transition probability for a single particle and then multiply this quantity by the difference in populations for the initial and final states. The effect being considered in the present work is an example of a process of a different kind in which the particle returns to the original level. In this case the dependence of the probability on the level populations is quadratic and the notion of a single-particle transition probability cannot be used.

An analysis of the general formula with multiparticle functions (such as Eq. (1) of [6]) leads to the following rule for processes in which particles are returned to an original level: The matrix element corresponding to each graph must be multiplied by the population of the initial (and final) states. For a process characterized by the absorption of a photon with frequency ω_1 and the simultaneous emission of photons at frequencies ω_2 and ω_3, we obtain the diagram shown in Fig. 1; this diagram yields the following expression for the probability:

$$2\pi t\delta(-\omega_1 + \omega_2 + \omega_3)\,|B_{31}^{(1)}\overset{+}{B}_{23}^{(2)}\overset{+}{B}_{12}^{(3)}|^2[N_1/(\omega_{31} - \omega_1)$$

$$\times (\omega_{21} - \omega_1 + \omega_2) + N_2/(\omega_{21} - \omega_3)(\omega_{23} - \omega_3 + \omega_1)$$

$$+N_3/(\omega_{32}-\omega_2)(\omega_{31}-\omega_2-\omega_3)]^2$$
$$=2\pi t\delta(-\omega_1+\omega_2+\omega_3)\,|B_{31}{}^{(1)}\overset{+}{B}_{23}{}^{(2)}\overset{+}{B}_{12}{}^{(3)}|^2$$
$$\times(\omega_{21}-\omega_3)^{-2}\left(\frac{N_1-N_3}{\omega_{31}-\omega_1}-\frac{N_2-N_3}{\omega_{32}-\omega_2}\right)^2 \qquad (1)$$

where B is the amplitude of the interaction operator and N_n is the population of the level.[1)] For reasons of simplicity the photon indices in (1) have been omitted.

The resonance singularities in (1) can be avoided in the usual way by introducing imaginary corrections to the characteristic frequencies; these correspond to the phenomenological introduction of relaxation parameters in the density-matrix equation. In general, however, this approach is not completely correct since it assumes a Lorentzian line shape. A somewhat better approach is to take account of the exact form of the interaction between the particles (cf. for example [6]). The singularity associated with the δ-function is avoided if damping is taken into account. For example, in a resonator the δ-function becomes a Lorentzian function which, upon exact tuning of the resonator to the frequency $\omega_3 = \omega_1 - \omega_2$, yields the factor $2Q/\pi\omega_3$ where Q is the loaded quality factor of the resonator.

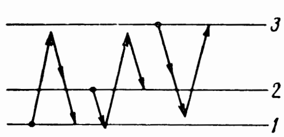

FIG. 1. Diagrams for computing three-photon transitions.

The probability for the present effect is equal to the difference between the expression in (1) and the probability for the inverse process, i.e., emission of a photon at frequency ω_1 with absorption of photons at frequencies ω_2 and ω_3. If we assume that the initial numbers of photons n_1, n_2, and n_3 at frequencies ω_1, ω_2, and ω_3 satisfy the inequality $n_1, n_2 \gg n_3$ the effect is proportional to $n_1(n_2+1)(n_3+1)-(n_1+1)n_2n_3 \approx n_1n_2$. Thus, to a first approximation the effect is independent of the strength of the field so long as the number of radiated photons is large. As expected, the final result agrees with that obtained by semiclassical methods.

[1)]It is assumed that the dimensions of the system are much smaller than a wavelength.

3. EXPERIMENT

The experimental investigation of frequency subtraction was carried out in a ruby single crystal with a volume of $0.15\ \mathrm{cm}^3$ and a 0.02% concentration of Cr ions. The three lower Zeeman levels were used and these are denoted in order of increasing energy by the numbers 1, 2, and 3. In order to resolve all three transitions the fixed magnetic field **H** is oriented perpendicularly to the axis of symmetry of the crystal ($\vartheta = 90°$). The dependence of the Bohr frequencies of ruby $\nu_{mn}(m,n = 1, 2, 3)$ on H for this orientation is shown in Fig. 2. It is evident from this figure that computing $\nu_{mn}(H)$ by means of the conventional spin-Hamiltonian for ruby leads to a considerable discrepancy with experiment. The experimental curves in Fig. 2 allow us to choose the parameters for a three-level ruby converter for the 3-centimeter region.

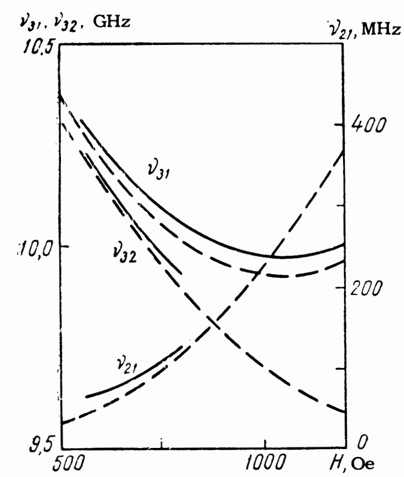

FIG. 2. Magnetic field dependence of the Bohr frequencies for the three lowest levels in ruby with the magnetic field H perpendicular to the crystal axis. The solid lines show the experimental results and the dashed lines show the results obtained with the spin Hamiltonian.

In the experiment the frequencies of the original fields ν_1 and ν_2 are 10.22 and 10.15 GHz so that with $H = 625\ \mathrm{Oe} \equiv H_0$ we have $\nu_1 = \nu_{31}$, $\nu_2 = \nu_{32}$ and $\nu_3 = \nu_{21} = 70$ MHz. For these values of ϑ and H the matrix elements for the components of the spin-operator S are $|S_{32}^{(x)}|^2 = |S_{31}^{(y)}|^2 = 0.95$, $|S_{21}^{(z)}|^2 = 2.2$ (the z-axis is parallel to the axis of symmetry of the crystal; the y-axis is parallel to H; the other components of S are much smaller than unity).

The crystal, of cylindrical shape, is located in a rectangular resonator in which the frequencies for the TE_{011} and TE_{101} modes are respectively

ν_1 and ν_2. The crystal is located at antinodes of the magnetic fields $\mathbf{H}_1$ and $\mathbf{H}_2$ where $\mathbf{H}_1 \parallel \mathbf{y}$ and $\mathbf{H}_2 \parallel \mathbf{x}$. The loaded quality factors for both modes are approximately 1000. Around the sample there is a detection coil with a quality factor of order 20 tuned to a frequency $\nu_3 = 70$ MHz and connected to a tuned amplifier. The axis of the coil is parallel to the x-axis.

The strength of the signal induced in the coil at the difference frequency is shown in Fig. 3a as a function of the field H (arbitrary units), where H varies from 440 to 880 Oe. The maximum power of the signal at the difference frequency P_3 is of the order of 2×10^{-14} W when the power of the input signals P_1 and P_2 is of the order of 50 mW. A theoretical estimate of the quantity P_3 can be obtained from the formula

$$P_3 = 8\pi^4 \nu_3 \frac{Q\varkappa}{V_3} (\Delta N)^2 \left(\frac{g\beta}{\hbar}\right)^6 |S_{32}{}^{(x)} S_{31}{}^{(y)} S_{21}{}^{(z)}|^2 \tau^4 H_1{}^2 H_2{}^2 f(H) \tag{2}$$

(here $\kappa \equiv H_3^2/\langle H_3^2\rangle$ and V_3 is the effective volume of the pickup coil; N is the number of Cr ions in the sample; $\Delta \equiv \rho_{33} - \rho_{17} \approx \rho_{33} - \rho_{22}$ is the difference in the diagonal elements of the density matrix; the spin-spin relaxation time τ is assumed to be the same in all transitions. This expression yields $P_3 \sim 10^{-14}$ W which is in good agreement with the experimental results.

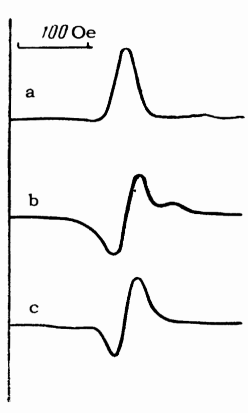

FIG. 3. The magnetic field dependence of the 70 MHz signal (a), the paramagnetic absorption at 10.15 GHz (b) and the paramagnetic absorption at 10.22 GHz (c).

The function f(H), which gives the line shape for the signal, is

$$f(H) = \left[1 + \left(\frac{H_0 - H}{\Delta H_{21}}\right)^2\right]^{-1} \left|\left(1 + i\frac{H_0 - H}{\Delta H_{31}}\right)^{-1} + \left(1 - i\frac{H_0 - H}{\Delta H_{32}}\right)^{-1}\right|^2, \tag{3}$$

where H_0 is the resonance value of the field for all three transitions and ΔH_{mn} is the line width. The experimental line width $(P_3)^{1/2}$ at the 0.7 level (cf. Fig. 3a) is $\Delta H = 24$ Oe. The curves in 3b and c represent the dependence on H of the paramagnetic absorption at frequencies ν_1 and ν_2 respectively as obtained by a double-modulation method. It follows from these results that $\Delta H_{31} = 39$ Oe, $\Delta H_{32} = 42$ Oe. If we assume that $\Delta H_{21} \gg \Delta H_{31} = \Delta H_{32} = 40.5$ Oe then according to Eq. (3) $\Delta H = \Delta H_{31}(2^{1/2} - 1)^{1/2} = 26$ Oe, which is in satisfactory agreement with the experimental value.

We have also investigated the dependence of the quantity P_3 on the angle ϑ. When ϑ varies within the limits $90° \pm 20'$ the quantity P_3 is reduced by a factor of four. This sharp dependence is determined by the function $S_{mn}(\vartheta)$ (especially $S_{21}^{(z)}(\vartheta)$).

The experimental results described here verify the basic theoretical predictions. As in the case of frequency addition (multiplication) (cf. for example, [5]) resonance between the frequencies of the field and the medium enhances considerably the effectiveness of the subtraction process. It should be noted that under the experimental conditions the spacing between levels 1 and 2 is of the same order as the width of the paramagnetic absorption line associated with the 3–1 and 3–2 transitions. At the same time Eq. (2) is based on the assumption that the initial frequencies ν_1 and ν_2 each excites its "own" transition independently: 1–3 or 2–3. Nonetheless, Eq. (2) yields satisfactory agreement with experiment because $\mathbf{H}_1 S_{32} \approx \mathbf{H}_2 S_{31} \approx 0$ for the chosen orientation of the magnetic fields with respect to the axis of symmetry of the crystal.[2])

The experiment reported here indicates the possibility of using three-level quantum-mechanical systems with discrete levels as resonance mixers for converting microwave signals into intermediate-frequency signals. In these experiments the conversion coefficient P_3/P_2 is 5×10^{-13} for heterodyne power $P_1 \sim 50$ mW. However, this quantity can evidently be increased considerably by optimization of the quantum-mechanical system (either solid or gaseous), by cooling the working material, and by using resonance systems with high quality factors.

[2])The small shoulder which appears in Fig. 3b to the right of the main line and in Fig. 3c to the left of the main line is due to the fact that these relations are starting to become exact equalities.

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