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Multiphoton transitions in gases at microwave frequencies

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Résumé. — Nous avons engendré des transitions à plusieurs photons en irradiant un gaz avec deux sources de radiation microonde. On a ainsi étudié des transitions à trois photons dans l'ammoniac et le sulphure de carbonyle en fonction de la fréquence, de la puissance rayonnée et de la densité des gaz. Nos résultats sont en assez bon accord avec les prédictions théoriques.

Abstract. — Multiphoton transitions are produced irradiating a gas by two microwave power sources. Three-photon transitions in NH_3 and OCS have been studied in function of frequencies, power and density of gases. Measurements agree fairly well with theoretical predictions.

Three-photon transitions in the optical region have been observed and related by some authors [1].

We describe a similar experiment that has been realized in the region of microwaves at centimetre wavelength in gases at few μHg of pressure.

The gas in a waveguide cell was irradiated by two strong waves at frequencies ν_1 and ν_2 , both very close either to the resonance frequency ν_0 of the (3,3) absorption line of inversion spectrum of NH_3 or to that of the line $J = 1 \rightarrow 2$ of pure rotational spectrum of carbonyl sulfide OCS. The frequency ν_1 is fixed at a distance $\Delta\nu$ from ν_0 , the second wave is swept in frequency between ν_0 and ν_1 . The frequency interval $\Delta\nu$ is several linewidth wide, and $\nu_1 < \text{or} > \nu_0$.

A schematic mechanism of three-photon transitions is shown in figure 1a and b. In the case $\nu_1 < \nu_0$ (Fig. 1a), the virtual levels are traced at a distance $\Delta\nu$ from real levels connected by the transition at ν_0 frequency. The process occurs when

$$2\nu_1 - \nu'_2 = \nu_0.$$

Two photons are absorbed from the wave ν_1 and one is emitted at the frequency ν'_2 . In the second case (Fig. 1b) the process is the same but the virtual levels are outside the real ones. The absorption and emission coefficients γ_1 and γ_2 have been calculated by means of the complex operator of Liouville [2]. The expansion parameters are of the kind

$$|E\mu/(\omega_0 - \omega_1)|$$

which are supposed to be < 1 , and in the hypothesis

$$|2\omega_1 - \omega_2 - \omega_0| \ll |\omega_1 - \omega_0|$$

Fig. 1a, b. — Energy level diagram : schematic of three-photon transition.

we have for the first case :

$$\gamma_1 = - \frac{8 \pi}{CE_1^2} \omega_1 NF^2 \frac{1/T_2}{(\omega_r - \omega_0 + S)^2 + 2 F^2 \frac{T_1}{T_2} + \frac{1}{T_2^2}} \frac{\omega_0 - \omega_2}{\omega_0 - \omega_1};$$

$$\gamma_2 = \frac{8 \pi}{CE_2^2} \omega_2 NF^2 \frac{1/T_2}{(\omega_r - \omega_0 + S)^2 + 2 F^2 \frac{T_1}{T_2} + \frac{1}{T_2^2}}$$

$$F = \mu^3 \frac{E_1^2 E_2}{(\omega_0 - \omega_1)(\omega_1 - \omega_2)}; \quad S = - \mu^2 \frac{2 E_1^2 + E_2^2}{\omega_1 - \omega_2}$$

with

T_1 : longitudinal relaxation time,

T_2 : transversal relaxation time (for ammonia we assume $T_1 \cong T_2$),

E_1, E_2 : electric fields of ν_1 and ν_2 waves,

μ : electric dipole moment of transition,

N : molecular density,

S : radiation shift,

and where $\omega_r = 2 \omega_1 - \omega_2$.

With appropriate approximations it is possible to deduce for the multiphoton an absorption of power of the kind [3] :

$$\Delta P = P_1 P_2^2 \quad (1)$$

and

$$\Delta P = (\omega_0 - \omega_1)^{-4} \quad (2)$$

and a dependence from the pressure equal to that of the one-photon line.

Figure 2 schematizes the spectrometer that we have set up to work in the K-band (24 GHz).

Two klystrons K_1 and K_2 are used as microwave generators (EM 1188) with 1 W of maximum power output. K_1 is stabilized in frequency through a tunable cavity C_1 with a very small frequency modulation at 5 kHz. The wave ν_2 is frequency modulated with the sawtooth of the oscilloscope. The waves ν_1 and ν_2 feed a Stark absorption cell, 50 cm length, through two decoupled arms of a magic T. The power of the two waves in the cell can be controlled and measured by means of two

Fig. 2. — Diagram of Stark spectrometer.

calibrated absorbers A_1 and A_2 . The signals are detected by the crystal X_2 in one of the H arms of a magic T, or by the crystal X_1 in the other H arm; in this case the signal is filtered by a tunable cavity C_2 .

These crystals are protected and insulated by means of a calibrated absorber A_3 and an uniline U_3 . The low-frequency signals are amplified by a lock-in and monitored on a scope.

The signals are optimized, when necessary, with the time-averaging technique and successively recorded. Since the two photons absorbed and the one emitted are of different frequency, the observation in X_2 evidences a net absorption while the detection in X_1 with the opportune tuning to ν_1 or ν_2 frequency of the cavity C_2 reveals the emission or the absorption (Fig. 3).

With A_1 and A_2 completely open (maximum power in the cell) a signal is detected in X_2 showing transitions at 1, 3 and 5 photons (Fig. 4).

The intensities of the three-photon transitions have been measured as a function of the power P_1 and P_2 . These measurements agree fairly well with (1) since it has been found a proportionality to

$$P_1^{0.96} \cdot P_2^{2.06}.$$

Fig. 3. — Emission (a) observed with the cavity C_2 tuned to the frequency ν_2 , and absorption (b) with the same cavity tuned to ν_1 i.e. the spectrometer is operating in the conditions reported in figure 1a.

Fig. 4. — A typical diagram showing absorption of one, three and five photons in NH_3 .

Fig. 5. — Three-photon line intensity as a function of the distance from the main (3,3) NH_3 absorption line.

At several values of P_1 and P_2 the intensity of three-photon line was measured point by point as a function of the frequency ν_1 , that is

$$\Delta\nu = \nu_0 - \nu_1$$

and it was found a dependence of the kind

$$\Delta P = \Delta\nu^{-4.2}$$

instead of a fourth-power behaviour (Fig. 5). The measurement reveals a dependence from the pressure of the linewidth which is the same and has the same value as that of the one-photon line.

In ammonia the effect is much stronger than in carbonyl sulphide and the measurements easier, but a good source of errors in the case of NH_3 is the instability of the pressure in the cell.

It is worthwhile to underline that amplification of a wave takes place, so that a tunable MASER can be conceived to work in the nearness of a microwave absorption line.

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