

Electric resonance in a gas

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Electric dipole resonance with selective absorption and dispersion at frequencies corresponding to the difference between the Stark sublevels is similar to the magnetic resonance at Zeeman-splitting frequencies, which is extensively used in radiospectroscopy. However, there is no known work on a direct realization of electric dipole resonance in a gas consisting of molecules with electric dipole moments. Such a realization is the content of the present communication.

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A gas placed in crossed electric fields, one constant E_0 and the other alternating $E \cos \Omega t$, undergoes a change $\Delta\epsilon$ in its permittivity at resonance with the transition frequency Ω . Calculation of the expected change of the permittivity can be carried out in the usual manner,^{1,2} and for a gas consisting of molecules of the symmetric-top type it leads to the expression

$$\Delta\epsilon_{\max} = \frac{\pi \Omega T_2 N_0 \frac{d^2}{3kT} \frac{K^2(2J+1)}{J(J+1)} \frac{4(4I^2+4I)}{4I^2+4I+1} \sqrt{\frac{B^2 C h^3}{\pi (kT)^3}}}{\sqrt{1 + T_1 T_2 \Omega^2 \left(\frac{E}{2E_0}\right)^2}} \quad (1)$$

Here T_1 and T_2 are the longitudinal and transverse relaxation times, N_0 is the molecule concentration, d is the dipole moment of the molecules, k is the Boltzmann constant, T is the absolute temperature, J is the rotational quantum number, K is the quantum number that determines the projection of the angular momentum on the molecule axis (it is assumed in (1) that K is not a multiple of three), I is the spin of the nuclei that do not lie on the molecule axis (these are assumed to be three identical nuclei), B and C are the rotational constants of the molecule, and h is Planck's constant.

When $\Delta\epsilon$ is determined from relation (1), for example for CH_3F molecules having $d = 1.79$ cgs esu under the condition $\Omega/2\pi = 25$ MHz, $K = J = 1$, $T_1 = T_2 = 10^{-7}$ sec, the gas pressure is 10^{-2} Torr and there is no saturation, the result at room temperature is $\Delta\epsilon_{\max} \approx 6 \times 10^{-10}$. If the gas is placed in a parallel-plate capacitor, then the relative change of the capacitance of the latter should also equal $\Delta c/c \approx 6 \times 10^{-10}$. Registration of such small capacitance changes is not a simple task, but is feasible. The published data on experiments with capacitance sensors for small displacements³ indicate that by now methods have been developed with which to measure capacitance changes $\Delta c/c \approx 10^{-11}$. Consequently, the electric dipole resonance can be experimentally registered.

For an experimental observation of the electric resonance we have assembled the

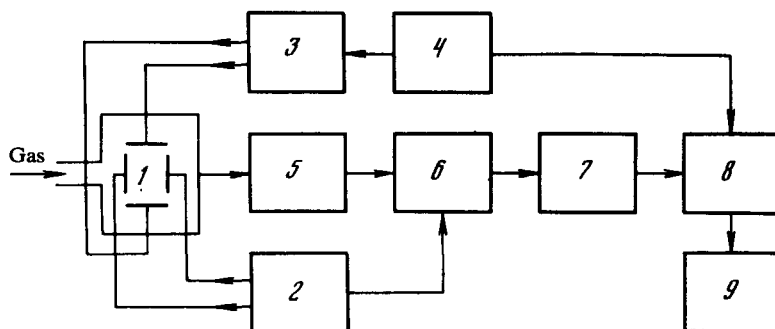


FIG. 1. Functional diagram of experimental setup: 1—gas cell, 2—25-MHz high-frequency generator, 3—dc source for the production of the field, 4—modulator, 5—high-frequency amplifier, 6—phase detector, 7—low-frequency amplifier, 8—synchronous detector, 9—automatic recorder.

setup whose functional diagram is shown in Fig. 1. The investigated gas fills, at a pressure 10^{-3} – 10^{-2} Torr, a gas cell with dimensions $30 \times 140 \times 360$ mm, in which homogeneous and mutually perpendicular constant and high-frequency electric fields are produced with the aid of two capacitor systems. The intensity of the constant field E_0 can be varied between 0 and 150 V/cm. The high-frequency field has a frequency 25 MHz and its intensity can be varied from 0 to 10 V/cm. The high-frequency capacitor is connected to an electronic circuit that can register small changes of the capacitance. The constant electric field in the gas cell is sawtooth-modulated with a period on the order of 5 minutes and is simultaneously harmonically modulated with frequency $\omega \approx 200$ Hz. The output of the measurement circuit, through a synchronous detector and an automatic recorder, is the amplitude A_2 of the second harmonic of the modulating harmonic signal. The installation can detect relative capacitance changes $\Delta c/c \approx 10^{-10}$. By measuring the phase of the reference voltage of the phase detector it is possible to register also the change of the absorption.

The energies of the Stark sublevels are given by¹

$$W_{JKM} = W_{JKM}^0 - E_0 d \frac{MK}{J(J+1)}. \quad (2)$$

Here W_{JKM}^0 is the energy of the (JKM) level in the absence of an electric field, and M is the quantum number that determines the projection of the kinetic angular momentum on the direction of the field E_0 . Taking into account the selection rule $\Delta M = \pm 1$, we obtain, with the aid of (2), the field intensity E_0 corresponding to resonance:

$$E_0 = \frac{\hbar J(J+1)\Omega}{dK}. \quad (3)$$

We see therefore that the smallest value of E_0 is obtained for $K = |J|$. In this case we get from (3)

$$E_0 = \frac{\hbar \Omega (J+1)}{d}. \quad (4)$$

Consequently, the values of E_0 corresponding to resonance at $J = 1, 2, \dots$ make up at $J = |K|$ an equidistant series. However, transitions with $|K| = J$ and $|K| \neq J$ are super-

imposed in such a way that only the first few resonances at the very smallest J can be observed without distortion.

The experiments were performed with CH_3F and CH_3CN . The CH_3CN molecule has a dipole moment $d = 3.92$ cgs esu. The nitrogen nucleus in this molecule has quadrupole moment. However, the quadrupole-splitting constant is small in this case and the quadrupole splitting turns out to be much less than the Stark splitting for the experimentally employed frequencies.

Figure 2 shows the plots for two different gases, CH_3F and CH_3CN , each plot

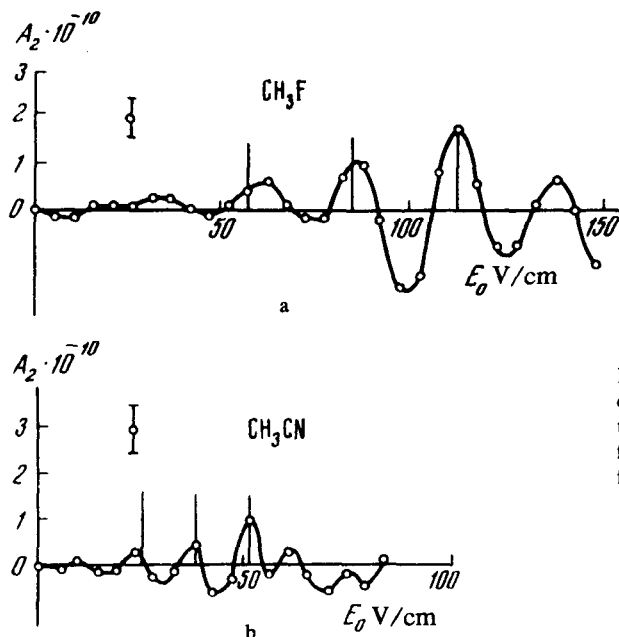


FIG. 2. Dependence of the amplitude of the second harmonic of the capacitance of the gas-cell capacitor on the field intensity E_0 : a—for CH_3F , b—for CH_3CN .

being an average of three traces of the recorder. The vertical lines in the same figure mark the values of E_0 that should correspond to resonances at $J = 1, 2$, and 3 in accordance with relation (4). It is seen from Fig. 2 that experiment actually reveals a number of resonance peaks whose positions correspond to relation (4). The growth of the amplitude of the resonance peaks with increasing J is in agreement with (1), if it is recognized that the first resonances take place at $|K| = J$.

This agreement between the calculation and the experimental results allows us to conclude that electric dipole resonance has been observed in the experiment and can be used in physical research.

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