Observation of a quantum-beat resonance between magnetic sublevels with $\Delta M = 4$

M. P. Auzin'sh and R. S. Ferber
P. Stuchka Latvian State University, Riga

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A nonlinear quantum-beat resonance between Zeeman sublevels differing by $\Delta M = 4$ has been observed experimentally. The measurements were taken in the $v' = 1, J'' = 72$ vibrational-rotational level of the $X\,^1\Sigma_g^+$ electronic ground state of the K_2 molecule. The Landé factor of the level has been determined.

The beat resonance, an extremely common type of interference between nondegenerate states,[1] was first detected in its nonlinear version during the optical orientation of atoms. The effect is seen as a change in the total absorption and fluorescence intensity when the frequency of the amplitude modulation of the pump light, ω_m, becomes equal to the magnetic-splitting frequency; this frequency is ω for a splitting of sublevels with $\Delta M = 1$ (orientation) or 2ω for a splitting of sublevels with $\Delta M = 2$ (alignment). The nonlinear nature of the optical pumping of the ground state, however, makes it possible to arrange a coherence of its magnetic sublevels with $\Delta M > 2$. A convenient method here is to perform the optical pumping by "emptying" diatomic molecules (Fig. 1), since polarization moments of order higher than the second arise in this case.[2,3] For example, if three absorption events occur while the coherence prevails in the ground state, a beat resonance between sublevels with $\Delta M = 4$ becomes possible. This letter reports the first observation of this effect.

During level crossing in a magnetic field $H = 0$ (the Hanle effect), ground-state polarization moments ϕ_q^κ of order $\kappa = 4$ and 6 appear.[2] Although the polarization

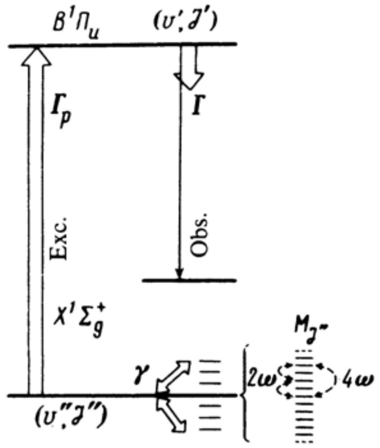

FIG. 1. Optical pumping through "emptying" during absorption of light in diatomic molecules.

moment with $\kappa = 6$, not $\kappa = 4$, produces the additional peak in the Hanle signal, it is ϕ_q^4, which is the highest-order moment, that is seen directly in the fluorescence, and its contribution to the signal is by no means small.[3] We can therefore expect to see a beat-resonance signal at the frequency $\omega_m = 4\omega$, in addition to the signals at ω and 2ω, which have been detected and studied previously in $^{130}Te_2$ molecules in the $X\,0_g^+$ electronic ground state ($v'' = 6$, $J'' = 52$).[4]

The present experiments were carried out on K_2 molecules. Linearly polarized light at 632.8 nm from a He-Ne laser excited the Q transition $X\,^1\Sigma_g^+(1.72) \rightarrow B\,^1\Pi_u(8.72)$ (Fig. 1). Under these conditions, absorption at a rate Γ_p competes with a radiative relaxation at a rate γ; the decay rate of the excited state is $\Gamma \gg \gamma \sim \Gamma_p$. The 60-mW beam from an LG-38 laser was sinusoidally modulated by an ML-102 electrooptic modulator with a modulation index $\epsilon \approx 0.8$. This modulation caused oscillations in the pumping rate: $\Gamma_p = \Gamma_{p0}(1 + \epsilon \cos \omega_m t)$. A cell filled with potassium vapor ($T = 460$ K; with an atomic number density $[K] = 0.7 \times 10^{14}$ cm^{-3} and a dimer number density $[K_2] = 0.7 \times 10^{14}$ cm^{-3}; see Ref. 5) was placed between the pole tips of an electromagnet which produced a field up to $H = 7200$ G (calibrated by an NMR method). The laser beam and its electric vector **E** were both perpendicular to **H**. The beam was passed back through the cell a second time by a mirror to improve the pumping efficiency. The fluorescence was observed along the **H** direction; for this purpose, a rotating mirror was placed between the pole tips of the magnet to direct the fluorescence to a DFS-12 monochromator (0.5 nm/mm), which singled out the wavelength corresponding to the transition $B\,^1\Pi_u(8.72) \rightarrow X\,^1\Sigma_g^+(16.72)$. It proved convenient experimentally to detect the nonlinear beat-resonance effect by measuring the time average of thc degree of linear polarization, $P = (I_\parallel - I_\perp)/(I_\parallel + I_\perp)$, a normalized quantity. For the measurements of P, the entrance slit was divided along its height into two parts by polarizers which transmitted the light ($I_\parallel$ and $I_\perp$) polarized parallel and perpendicular to the vector **E**. The corresponding light beams at the exit slit were directed by lightguides to two FÉU-79 photomultipliers and were ultimately measured in a two-channel photon-counting system.

We measured the dependence of P on the modulation frequency ω_m at a fixed magnetic field (6750 G in the case in Fig. 2). To eliminate any drift effect, we measured

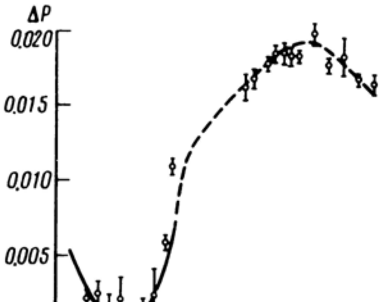

FIG. 2. Degree of polarization of the fluorescence vs the frequency at which the exciting light is modulated.

P at a selected reference frequency, ω_m^v (the double circle) before and after each measurement of $P(\omega_m)$, integrating the signal over 1 min (a count rate $\sim 10^4$ counts/s). The difference $P(\omega_m) - P(\omega_m^0) = \Delta P$ is shown in Fig. 2. The total time required to measure the results in Fig. 2 was about 3 h; the various points were measured in random order.

We see two resonances in Fig. 2. The first is at a frequency of about 200 kHz and agrees both in position and in the sign of the effect[4] with a beat resonance between Zeeman sublevels of the ground state with $\Delta M = 2$; i.e., this frequency is 2ω, when we set the Landé factor $g = 1.177 \times 10^{-5}$, which was found in Ref. 6 for the entire thermal-equilibrium set of levels (v'', J'') of the $(X\,^1\Sigma_g^+)$ state of K_2. The second resonance, at a frequency of about 400 kHz, is of the opposite sign and corresponds to the expected beat resonance between sublevels with $\Delta M = 4$ at the frequency 4ω. Experiments in a stronger field, $H = 7150$ G, yielded the same result, with the expected frequency shift of the beat resonance. We were prevented from varying H over a broader range because the signal quality (the signal-to-width ratio) became unacceptably poor at lower magnetic fields, while the capabilities of the magnet set an upper limit on the field. The beat resonance at 2ω can be described quite simply by solving the system of equations for the polarization moments through a series expansion and retaining the term proportional to Γ_{p0}^2/γ^2 (Ref. 4). The time-average degree of polarization $\overset{(2)}{P}$ for Q transitions in this approximation is[7] (we are also assuming that the relaxation rate of the polarization moments does not depend on the order of the moments)

$$\overset{(2)}{P} = \frac{0.5}{1 + 4\,\Omega^2/\Gamma^2}\left[1 - \frac{3\Gamma_{p0}(D_{22} - 8E_{22})}{14 - 3\Gamma_{p0}\,(2D_{20} + D_{22})}\right], \tag{1}$$

where

$$D_{\kappa q} = 0.5\,(B_{\kappa q} + B_{\kappa\,-q}), \tag{2}$$

$$E_{\kappa q} = \frac{\Omega}{2qi\Gamma}\,(B_{\kappa q} - B_{\kappa\,-q}), \tag{3}$$

$$B_{\kappa q} = \frac{\gamma + iq\omega}{\gamma^2 + q^2\omega^2} + 0.5\,\epsilon^2\,\frac{[(\gamma + iq\omega)^2 + \omega_m^2](\gamma - iq\omega)}{[\gamma^2 + (\omega_m - q\omega)^2][\gamma^2 + (\omega_m + q\omega)^2]}\,. \tag{4}$$

Here $\Omega = g_{J'}\mu_0 H/\hbar$ is the Zeeman frequency for the $B\,^1\Pi_u$ state, $g_{J'} = 1/J'(J'+1)$, μ_0 is the Bohr magneton, and, correspondingly, $\omega = g_{J''}\mu_0 H/\hbar$. Equations (1)–(4) incorporate polarization moments of order no higher than $\kappa = 2$, and the beat resonance is related to the decrease in $\overset{(2)}{P}$ caused by the restoration of ϕ_2^2. In order to describe the beat resonance at 4ω in this manner we need to go to at least the next approximation; i.e., we need to incorporate the polarization moments with $\kappa = 4$, where the effect begins to be seen. These calculations have not yet been carried out. It nevertheless follows from an analysis of the solution of the system of polarization moments for the case of the Hanle effect with steady-state excitation[2] that incorporating ϕ_q^4 leads to an increase in P at a fixed H, so that we would expect the beat-resonance signal at $\omega_m = 4\omega$ to have the sign opposite that at 2ω, as we in fact observe experimentally.

Variation of the parameters $g_{J''}$, γ, and Γ_{p0} in (1)–(4) has made it possible to find the Landé factor for the isolated level $v'' = 1$, $J'' = 72$ of the $(X\,^1\Sigma_g^+)$ diamagnetic state of K_2 for the first time, through an analysis of the signal at 2ω (the solid curve in Fig. 2). The average value over all the experiments is $g_{J''} = (0.99 \pm 0.04) \times 10^{-5}$. With the constant values $\Gamma = 8.62 \times 10^7\ s^{-1}$ and $\gamma = \Gamma_{p0} = 1.2 \times 10^6\ s^{-1}$ in (1)–(4), the minimum is shifted to the right of $\omega_m = 2\omega$ to about 2.075ω by the Hanle effect of the excited state. The 4ω resonance may also exhibit an additional shift, in the same direction, because of the "substrate" of the 2ω signal. Since the signal shape has not been calculated, we did not use the 4ω beat-resonance effect to determine $g_{J''}$, although in principle the resonances at high frequencies may hold promise because of the better ratio of the frequency of the signal to its width.

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