

DETERMINATION OF THE GROUND-STATE LANDÉ FACTOR FOR DIATOMIC MOLECULES BY A BEAT-RESONANCE METHOD

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Received 22 July 1989; in final form 2 January 1990

Non-linear resonance of quantum beats under harmonically modulated excitation in a magnetic field is considered as a method of determining the Landé factor for a single electronic-vibrational-rotational (EVR) level of a diatomic molecule in its electronic ground state. The role of the sign of the Landé factor in the ground and excited levels, stimulated emission and "fly-through" relaxation conditions are analysed using a semiclassical description of the beat resonance signal in laser-induced fluorescence. Refined Landé factor values of EVR levels of $^{39}\text{K}_2(X^1\Sigma_g^+)$ and $^{130}\text{Te}_2(XO_g^+)$ are obtained by means of fitting experimental signals.

1. Introduction

As a rule, the orbital and spin angular momenta of diatomic molecules in the ground state have zero-value projection on the molecular axis. In this case magnetism may be due to nuclear rotation, as well as to non-adiabatic term interaction (the "electronic" contribution). In this connection experimental data on Landé g factors of separate rovibronic levels are of considerable interest. g factor measurements of such states are based on the optical pumping effect [1] (in the sense of orientation or alignment) of the lower EVR level in the process of its depopulation, e.g. in the process of optical absorption. During absorption, coherence between magnetic sublevels of the ground state is created, which is transferred by the same light field to the excited state and manifests itself through subsequent fluorescence. Coherence may be destroyed by an external magnetic field causing a splitting of the degeneracy of magnetic sublevels (ground-state level crossing effect [2]), but may also be restored in case of modulated pumping [3], producing quantum beats after pulsed excitation and beat resonance (BR) at harmonically modulated excitation. The BR method can be easily effected at laser excitation, see for example ref. [4] for neon.

The methods have first been applied to ground-state molecules in refs. [2,5,6] for Te_2 and K_2 . Correct description of the shape of the registered non-linear signal is required for precise determination of the splitting frequency, accounting for resonance peak shifts in the presence of the pumping light field. From this point of view the approximation applied in ref. [5] and giving an analytical description in terms of polarization moments is not sufficiently precise. This fault is removed in refs. [7,8] by an approach based on classical description of angular momentum orientation. The present paper forms a further stage in this approach. It takes into account effect of stimulated radiation, fly-through effects, and also analyses the role of Landé factor sign of states interconnected by laser radiation. This has made it possible to obtain refined Landé factor values of rovibronic levels of $^{39}\text{K}_2(X^1\Sigma_g^+)$ and $^{130}\text{Te}_2(XO_g^+)$.

2. General description of BR signals

Experimental methods and techniques are described in sufficient detail in refs. [5,8]. Laser radiation, amplitude-modulated at frequency ω causes a rovibronic transition $(\alpha'', v''_a, J''_a) \rightarrow (\alpha', v'_b, J'_b)$ or $a \rightarrow b$, fig. 1a, at a rate (in approximation of a broad excitation line) $\Gamma_p = \Gamma_{p0}(1 + \epsilon \cos \omega t)$, ϵ being the modulation depth. The glass cell containing vapour of the test element (connected with a vacuum system) is placed in the magnetic field \mathbf{B} ; the direction of \mathbf{B} , of excitation and of observation ($b \rightarrow c$ transition) at various polarizations are shown in figs. 1b and 1c. In experiments at constant magnetic field \mathbf{B} the modulation-frequency ω dependence of the normalized value of $(I_1 - I_2)/(I_1 + I_2)$ was registered. Here I_1 and I_2 are either two linearly polarized components of fluorescence intensity at linearly polarized light excitation (fig. 1b), or two circularly polarized components at circularly polarized excitation (fig. 1c). In other words, we have here conventional measurements of the degree of linear polarization P , or of circular polarization C . In all cases I_1 and I_2 are time-averaged radiation intensities. Typical BR signals are presented in figs. 2 and 3.

In describing the BR effect we shall base ourselves on classical equations of motion, as presented and discussed in refs. [9,10] and valid for $J', J'' \gg 1$, for probability density $\rho_\alpha(\theta, \varphi)$, where α denotes states a or b, θ, φ are spherical angles describing rotational momentum orientation, the z axis being directed along the magnetic field \mathbf{B}

$$\frac{\partial}{\partial t} \rho_a = -\Gamma_a \rho_a - \mathbf{u} \frac{\partial \rho_a}{\partial \mathbf{r}} - \omega_a \frac{\partial \rho_a}{\partial \varphi} + \Gamma_p C_{\theta, \varphi} (\rho_b - \rho_a) + \Gamma_{ba} \rho_b + \lambda_a, \quad (1a)$$

$$\frac{\partial}{\partial t} \rho_b = -\Gamma_b \rho_b - \mathbf{u} \frac{\partial \rho_b}{\partial \mathbf{r}} - \omega_b \frac{\partial \rho_b}{\partial \varphi} - \Gamma_p C_{\theta, \varphi} (\rho_b - \rho_a). \quad (1b)$$

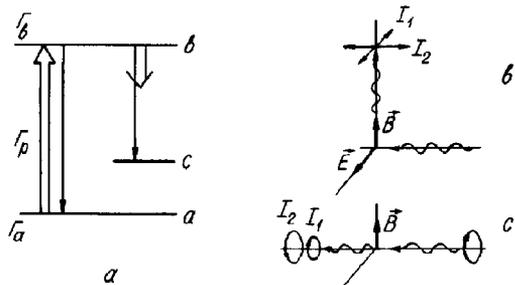


Fig. 1. Scheme of transitions (a) and of geometry of experiment (b) and (c) producing optical alignment or orientation of lower level a.

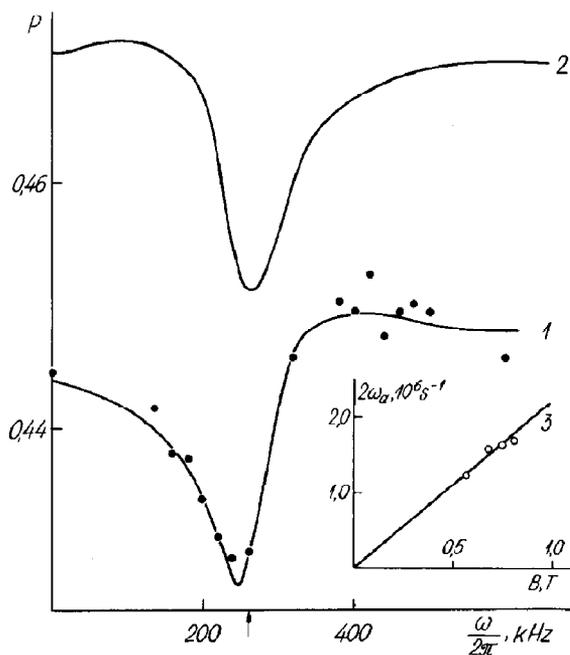


Fig. 2. Experimental points of BR signal for $^{39}\text{K}_2$ ($X^1\Sigma_g^+, 1, 73$) at $B=0.743$ T and their approximation by calculated curve (1); the calculated curve, as obtained at the same parameter, but at equal signs of g_a and g_b (2).

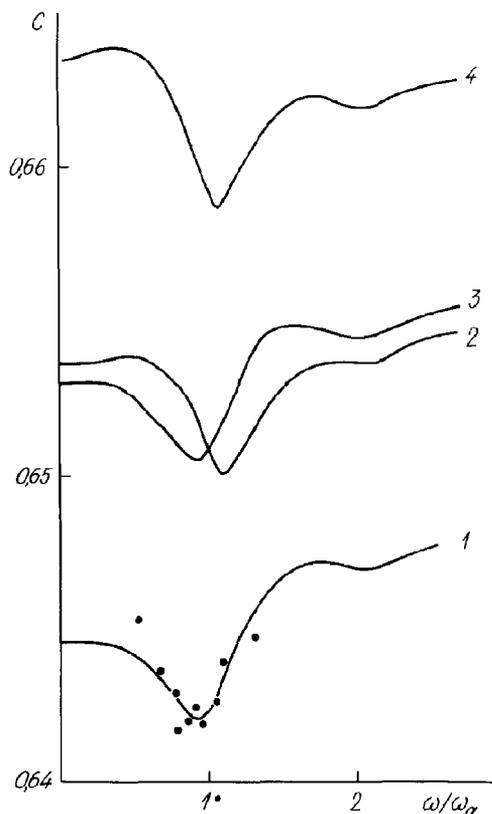


Fig. 3. Experimental points [5] of BR signal at orientation upon $^{130}\text{Te}_2$ ($X0_0^+$, 6, 52) with $B=0.0926$ T and their approximation by calculated curve (1) at $\Gamma_{p0}=0.15 \times 10^6 \text{ s}^{-1}$; $\Gamma_b=1.94 \times 10^6 \text{ s}^{-1}$; $\Gamma_a^{\Sigma}=0.41 \times 10^6 \text{ s}^{-1}$; $\omega_a=1.68 \times 10^6 \text{ s}^{-1}$. Curve (2) is obtained without account for different signs of g_a, g_b ; curves (3) and (4) are obtained neglecting stimulated radiation.

Here $C_{\theta,\varphi}$ is the coefficient of angular dependence of absorption probability, the terms with $\omega_{\alpha}=g_{\alpha}B\mu_0/\hbar$ describe the precession of the angular momentum J_{α} around B , terms with Γ_p the absorption and stimulated emission, and with Γ_{ba} the spontaneous transitions. Summands containing velocities u and coordinates r take into account radiationless relaxation due to thermal molecular motion through the laser beam. The term $\Gamma_a\rho_a$ describes collisional depopulation, but λ_a is the repopulation of the ground state; Γ_b is the effective relaxation rate of the excited state.

Let us divide the discussion of manifestation of BR into two parts. We shall first neglect the r and u dependence, which would apply for the case of purely collisional relaxation. Under experimental conditions with K_2 and Te_2 (cf. table 1) this does not take place at the electronic ground state. Indeed, the reciprocal value of transit time of particles through the laser beam of 2–3 mm diameter is comparable to the rate of collisional relaxation $\sigma_{\text{col}}v_{\text{rel}}N$, v_{rel} being the average relative velocity, N the concentration. We may nevertheless expect, as follows from the discussion presented in ref. [11], that the description of collisions and transit by one summary constant Γ_a^{Σ} ("collisional" account of transit) will not cause too large an error at low values of the pumping parameter $\Gamma_{p0}/\Gamma_a^{\Sigma} \approx 1$, cf. table 1. In this approximation, when Γ_a is replaced by Γ_a^{Σ} in (1a), and $\partial\rho_{\alpha}/\partial r=0$, the system (1) is solved numerically, applying a method similar to that used in ref. [8], viz. by expanding ρ_{α} into a two-dimensional Fourier series over φ and ωt ,

$$\rho_{\alpha} = \sum_{kl} \rho_{\alpha}^{kl}(\theta) \exp(ik\varphi + i\omega t), \quad \alpha = a, b. \quad (2)$$

The g_{α} values for the above dimers are then obtained by fitting theoretical P or C values (see below). Next, we shall attempt to proceed further in the discussion of the BR signal for the case when we have only fly-through relaxation introducing a number of other simplifying assumptions.

Table 1

Basic parameters of the experiment and the combining EVR levels interconnected by the laser transition

Parameters	$^{39}\text{K}_2$	$^{130}\text{Te}_2$
λ_{exc} (nm)	632.8	514.5
a or (α'' , ν''_a , J''_a)	(X $^1\Sigma_g^+$, 1, 73)	(X 0_g^+ , 6, 52)
b or (α' , ν'_b , J'_b)	(B $^1\Pi_u$, 8, 73)	(A 0_u^+ , 11, 53)
T (K)	441	630
N (cm^{-3})	[K] = 0.3×10^{14} [12]	[Te ₂] = 0.8×10^{14} [13]
Γ_a^Σ (10^6 s^{-1})	0.13+0.23	0.39+0.41
σ_{col}^a (10^{-14} cm^2)	3.3 for (K ₂ +K) [11]	4 for (Te ₂ +Te ₂) [14]
Γ_b (10^6 s^{-1})	86.2 [2]	1.49 [14]
Γ_{p0} (10^6 s^{-1})	0.22+0.35	0.16+0.12
ϵ	0.8	0.8
Γ_{ba} (10^6 s^{-1})	3.78 [15]	0.037 [16]
g_b (10^{-4})	$-1/J'(J'+1) = -1.85$	0.52 [14]
g_a (10^{-4})	0.124 ± 0.007^a	-1.96 ± 0.08^a

^{a)} This work.

3. $^{39}\text{K}_2$ molecule

In the case of K₂ a Q-type transition has been effected in experiments of this type [6,8,11], cf. table 1, applying linearly polarized light. Hence, we shall be interested in the observed degree of polarization *P* in this case. According to the definition, in the case of Q transitions ($J''_a = J'_b = J''_c$), the formula

$$P = \int \langle \rho_b \rangle_{r,u,t} \sin^2 \theta \cos 2\varphi \, d \cos \theta \, d\varphi \bigg/ \int \langle \rho_b \rangle_{r,u,t} \sin^2 \theta \, d \cos \theta \, d\varphi \quad (3)$$

holds, $\langle \rangle_{r,u,t}$ denoting averaging over variables according to respective indices. It should be noted that $C_{\theta,\varphi} = \sin^2 \theta \cos^2 \varphi$ in (1). In order to find $\langle \rho_b \rangle$ the expansion (2) was substituted into (1), and then a system of algebraic equations was solved for $\rho_{\alpha'}^{k,l}$. It was taken into account that owing to averaging, only harmonics with $l=0$ contribute to (3). Sufficient accuracy is achieved at $|k, l| \leq 10$, no terms with odd k, l values appearing. As can be seen from data for K₂, cf. table 1, stimulated radiational transitions are negligibly small owing to $\Gamma_{p0}/\Gamma_b \approx 10^{-2}$. In this case eq. (1a) is solved independently and the $\rho_{\alpha'}^{k,l}(\theta)$ values obtained are then substituted into (1b), in order to calculate the necessary $\rho_b^{k,0}(\theta)$ values.

Fig. 2 shows experimental points for K₂. In the approximation process parameters Γ_a^Σ , Γ_{p0} , g_a were varied; the other parameters entering into (1) are presented in table 1. The fitting results are as follows: $\Gamma_a^\Sigma = 0.15 \times 10^6 \text{ s}^{-1}$, $\Gamma_{p0} = 0.35 \times 10^6 \text{ s}^{-1}$, $g_a = 1.24 \times 10^{-5}$. The Zeeman splitting between magnetic sublevels with $|\Delta M''| = 2$ equals $2\omega_a/2\pi = 257 \text{ kHz}$ (arrow in fig. 2). It is essential that the calculated curve depends on the sign relations of g_a and g_b , although it may be assumed in the given experimental geometry that the signal does not depend on the sign of the *g* factor. Curve (1) was obtained taking into account different signs of Landé factors, since $g_a > 0$ and $g_b < 0$, as experimentally confirmed in ref. [17]. The positive sign of g_a here is due to the dominant contribution of rotating nuclei, analogous to the case of molecular hydrogen, see refs. [18,19]. The influence of the nuclear spins in $^{39}\text{K}_2$ should be negligible because the abovementioned Zeeman splitting value between $|\Delta M''| = 2$ is much larger than the hyperfine splitting. Indeed the latter does not exceed 3 kHz if we use the electric quadrupole interaction constant value [18] $eqQ = 158 \text{ kHz}$ (for $J''_a = 73$). The contribution of other interactions is much smaller, cf. recent results for $^{23}\text{Na}_2$ in ref. [20]. Curve (2) demonstrates calculation results with the same parameters, but for equal signs of g_a, g_b . It may be seen that this faulty account of the g_a and g_b sign relations leads, firstly, to certain divergence in the determined value of $2\omega_a$ (and hence also in g_a).

Secondly, there is considerable change in the range of P values and in the shape of the whole curve. In particular, the small "hump" near 400 kHz disappears which is, most likely, connected with BR between magnetic sublevels with $|\Delta M''|=4$ and may be detected experimentally in more meticulous measurements [21].

We have also reconsidered the previous experimental data as obtained in ref. [8], taking into consideration the signs of Landé factors. Linear approximation for the obtained B dependence of $2\omega_a$, cf. straight line (3) in fig. 2, yields $g_a = (1.24 \pm 0.07) \times 10^{-5}$, the error corresponds to a reliability of 0.95. This value, refined as compared to $g_a = (1.01 \pm 0.04) \times 10^{-5}$ [8]^{#1}, is in better agreement with that obtained from quantum beats in kinetics [22], namely $g_a = 1.30 \pm 0.27$. Incidentally, magnetic resonance in molecular beams without rovibronic level selection yielded a value of 1.18×10^{-5} [18].

4. $^{130}\text{Te}_2$ molecule

In this case transitions of P-, R-type have been produced. This permitted effective optical orientation of Te_2 dimers at circularly polarized excitation, cf. fig. 1. In the experiment the degree of circularity C was determined, which can be expressed through components $\rho_b^{k,l}(\theta)$ entering into (2), namely

$$C = \frac{\int 4 \operatorname{Im} \rho_b^{1,0}(\theta) \sin \theta \, d \cos \theta}{\int [2 \rho_b^{0,0}(\theta) (1 + \frac{1}{2} \sin^2 \theta) - \operatorname{Re} \rho_b^{2,0}(\theta) \sin^2 \theta] \, d \cos \theta}. \quad (4)$$

Another peculiarity consists in a considerable increase in the contribution of stimulated radiation, cf. table 1. If that is allowed for, eqs. (1) are solved simultaneously. Previously [5] obtained BR signals in $\text{Te}_2(\text{X}0_g^+)$ orientation have now been reprocessed by us allowing for induced transitions, as well as for different signs of g_a and g_b [17], cf. fig. 3, curve (1). Apart from BR at $|\Delta M''|=1$ in the form of a minimum in the vicinity of $\omega/\omega_a=1$, another small minimum has also been observed [5] near $\omega/\omega_a=2$, due to restored alignment. Curve (3) was calculated for the same conditions, but without accounting for stimulated transitions, whilst curves (2) and (4) – for equal signs of g_a and g_b , respectively with and without accounting for stimulated radiation. Their comparison demonstrates once again the importance of allowing for the signs of g_a . On disregarding stimulated transitions (here $\Gamma_{p0}/\Gamma_b \approx 0.1$, i.e. their part is small), we obtain signals of slightly different shape and a very small shift (not visible in the scale of fig. 3) of the minimum towards higher values of ω/ω_a . The B dependence of ω_a within the 0.1–0.2 T range of B , as treated on the basis of experimental data in ref. [5] after a new method, yields a value of $g_a = -(1.96 \pm 0.08) \times 10^{-4}$ for $^{130}\text{Te}_2(\text{X}0_g^+, 6, 52)$, differing from that in ref. [5], namely $g_a = -(1.68 \pm 0.05) \times 10^{-4}$. It may be of interest to note that the calculation according to formula $g_a = -4B_e/\lambda_{10}$ [23], where $B_e = 0.03967 \text{ cm}^{-1}$ is the rotational constant, and $2\lambda_{10} = 1975 \text{ cm}^{-1}$ [24] is Ω , the doubling between $\text{X}0_g^+$ and $\text{X}1_g^+$ states, yields a value of $g_a = -1.61 \times 10^{-4}$. It is not impossible that an excess of 20 pc of the experimental value above the previously mentioned theoretical value may be due to the effect of higher situated terms.

5. BR signals under conditions of fly-through relaxation

Let us now consider system (1) under conditions when in (1a), not the first but the second relaxation term, which contains coordinate and velocity dependence and describes fly-through relaxation, is dominant. Let us assume the following simplifications: let $\Gamma_a=0$ and $\Gamma_{ba}=0$; we shall neglect the term $u \partial \rho / \partial r$ in comparison to $\Gamma_b \rho_b$; in addition, we shall consider a one-dimensional problem with motion proceeding along the x axis. We

^{#1} Regrettably, in fig. 2b of ref. [8] the indicated values of both axes should actually be 10 times smaller.

multiply both equations obtained by an arbitrary, but periodic (with period 2π) function $A(\varphi)$. Adding and averaging we obtain

$$-\omega_b \left\langle \rho_b \frac{dA}{d\varphi} \right\rangle_{\varphi x u t} + \langle u A \rho_b \rangle_{\varphi u t} \Big|_{x_0}^{x_1} + \Gamma_b \langle A \rho_b \rangle_{\varphi x u t} - \omega_a \left\langle \rho_a \frac{dA}{d\varphi} \right\rangle_{\varphi x u t} + \langle u A \rho_a \rangle_{\varphi u t} \Big|_{x_0}^{x_1} + \Gamma_a \langle A (\rho_a - \rho_a^0) \rangle_{\varphi x u t} = 0, \quad (5)$$

where x_0, x_1 are the "entering" and "leaving" coordinates of the molecule in the space of the laser beam, the profile of which we assume as being rectangular. The radiation completely "pumps out" level a, so that at $x = x_1$ we have $\rho_a = 0$, and at $x = x_0$ we have $\rho_a = \rho_a^0, \rho_b = 0$. Let us consider expression (3) for a degree of polarization P . Assuming $A = 1, A = \cos 2\varphi$ and $A = \sin 2\varphi$ in (5), we obtain

$$\langle \rho_b \rangle_{\varphi x u t} = \frac{1}{\Gamma_b} \langle u \rho_a^0 \rangle_u, \quad (6)$$

$$\langle \cos(2\varphi) \rho_b \rangle_{\varphi x u t} = - \frac{2\omega_a/\Gamma_b}{1 + 4\omega_b^2/\Gamma_b^2} \left\langle \left(\sin 2\varphi + \frac{2\omega_b}{\Gamma_b} \cos 2\varphi \right) \rho_a \right\rangle_{\varphi x u t}. \quad (7)$$

It may be seen that the denominator (3) does not depend on modulation frequency ω and on magnetic field, so that the BR signal in $P(\omega)$ is completely determined by the numerator, i.e. by expression (7). We now introduce the following notation:

$$G = G_1 + 2 \frac{\omega_b}{\Gamma_b} G_2, \quad G_1 = - \langle \sin(2\varphi) \rho_a \rangle_{\varphi x u t}, \quad G_2 = - \langle \cos(2\varphi) \rho_a \rangle_{\varphi x u t}. \quad (8)$$

Let the excitation intensity be homogeneous at $x > x_0$; for simplicity we put $x_0 = 0$. Then the solution for $\rho_a(\varphi, x, u, t)$ may be represented in the form

$$\rho_a(\varphi, x, u, t) = \exp \left[- \frac{1}{u} \int_0^x dx_1 \Gamma_p \left(t - \frac{x_1}{u}, \varphi - \omega_a \frac{x_1}{u} \right) \right], \quad (9)$$

where $\Gamma_p(t, \varphi) = \Gamma_{p0} (1 + \epsilon \cos \omega t) \cos^2 \varphi$. Averaging over φ for $\tilde{G}_1 = \frac{1}{2} \Gamma_{p0} G_1, \tilde{G}_2 = \frac{1}{2} \Gamma_{p0} G_2$, we obtain

$$\begin{aligned} \tilde{G}_1 &= \frac{1}{2\pi} \int_0^{2\pi} d\tilde{t} \int_0^\infty d\tilde{\tau} \frac{e I_1(\sqrt{d^2 + e^2})}{\sqrt{d^2 + e^2}} \exp(-f), & \tilde{G}_2 &= \frac{1}{2\pi} \int_0^{2\pi} d\tilde{t} \int_0^\infty d\tilde{\tau} \frac{d I_1(\sqrt{d^2 + e^2})}{\sqrt{d^2 + e^2}} \exp(-f), \\ d &= \int_0^{\tilde{\tau}} d\tilde{\tau}_1 (1 + \epsilon \cos(\tilde{t} - \tilde{\omega} \tilde{\tau}_1) \cos(2\tilde{\omega}_a \tilde{\tau}_1)), & e &= \int_0^{\tilde{\tau}} d\tilde{\tau}_1 (1 + \epsilon \cos(\tilde{t} - \tilde{\omega} \tilde{\tau}_1) \sin(2\tilde{\omega}_a \tilde{\tau}_1)), \\ f &= \int_0^{\tilde{\tau}} d\tilde{\tau}_1 (1 + \epsilon \cos(\tilde{t} - \tilde{\omega} \tilde{\tau}_1)), \end{aligned} \quad (10)$$

where $\tilde{\tau} = (x/u)(\frac{1}{2}\Gamma_{p0})$, $\tilde{t} = \omega t$, $\tilde{\omega} = 2\omega/\Gamma_{p0}$, $\tilde{\omega}_a = 2\omega_a/\Gamma_{p0}$, I_1 is a modified Bessel function. At linear polarization and for Q transition we have $\Gamma_{p0} = \Gamma_{p00} \sin^2 \theta$. Radiation intensities I_1 and I_2 are also proportional to $\sin^2 \theta$. Hence,

$$P \approx \frac{\omega_a}{\Gamma_{p00}} \frac{1}{1 + 4\omega_b^2/\Gamma_b^2} \int d \cos \theta \left(\tilde{G}_1 + 2 \frac{\omega_b}{\Gamma_b} \tilde{G}_2 \right). \quad (11)$$

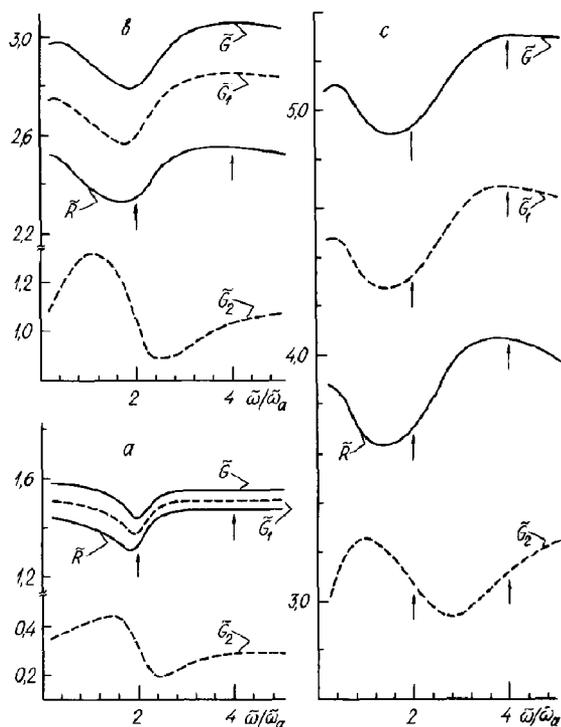


Fig. 4. ω/ω_a dependence of magnitudes \tilde{G}_1 , \tilde{G}_2 , \tilde{G} , \tilde{R} determining the BR signal in expression (11) for $\tilde{\omega}_a = 2\omega_a/\Gamma_{p0}$, $\tilde{\omega}_a = 2.4$ (a), $\tilde{\omega}_a = 1.2$ (b) and $\tilde{\omega}_a = 0.6$ (c).

The above transition may be useful in the analysis of experimental results. Indeed from (10) follows automodelity of \tilde{G}_1 and \tilde{G}_2 over the variable $\tilde{\omega}_a$. This means that at simultaneous change by the same number of times of the intensities of the excitation light and of magnetic field the \tilde{G}_1 and \tilde{G}_2 values do not change. This may lead to definite conclusions on the behaviour of P at such a change, cf. (11). Thus, for $\omega_b = 0$, at change of magnetic field and excitation intensity m times the value of P will not change, whilst the resonance frequency will increase m times.

Fig. 4 shows calculated curves for \tilde{G}_1 , \tilde{G}_2 , $\tilde{G} = \tilde{G}_1 + (2|\omega_b|/\Gamma_b)\tilde{G}_2$, $\tilde{R} = \tilde{G}_1 - (2|\omega_b|/\Gamma_b)\tilde{G}_2$ at different values of $\tilde{\omega}_a$. It may be seen that \tilde{G}_1 and \tilde{G}_2 oscillate in antiphase. As a result the position of resonance for \tilde{R} (opposite signs of magnetic splitting ω_a and ω_b) lies further to the left than for \tilde{G} (equal signs). The position of resonance is close to $\omega = 2\omega_a$ only at low pumping efficiency $\Gamma_{p0}/\Gamma_a \ll 1$ (corresponds to higher $\tilde{\omega}_a$ values); it shifts to the left with increase in pumping rate Γ_{p0} . Resonance near frequency $\omega = 4\omega_a$ becomes noticeable only at high pumping levels and is considerably more pronounced for \tilde{R} than for \tilde{G} . Thus, under fly-through conditions the behaviour of $P(\omega)$ shows all the same qualitative peculiarities as under conditions of distinct collisional relaxation, cf. fig. 2 and also curves presented in ref. [8].

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