Nonlinear resonance in low-pressure gases not subject to drift broadening

A. M. Shalagin

Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR

(Submitted 30 June 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **34**, No. 4, 193–196 (20 August 1981)

The higher-order nonlinear-absorption resonances with respect to the radiation intensity are not subject to a drift broadening (broadening attributable to the finite time the absorbing particle spends in the light beam). This effect is due to the increased role of slow particles resulting from the increase of the degree of interaction nonlinearity. Starting with the third-order intensity, the nonlinear resonances retain the collision width at any low pressure.

PACS numbers: 51.70. + f

Nonlinear resonances of minimum width must be produced for nonlinear ultra-high-resolution spectroscopy, in particular, for building lasers with a highly stabilized emission frequency. Long-lived atomic or molecular systems under low-pressure conditions are used for this purpose. The pressure decrease is dictated by the need to reduce collision broadening. Unfortunately, unlimited narrowing of the nonlinear resonance cannot be achieved by reducing the pressure, because the so-called drift-broadening effects, which are attributable to the finite time the particle spends in the light beam, become a factor. At present, this factor is restricting further progress in the experimental achievement of ultranarrow nonlinear resonances.[1-3]

It has been shown[4,5] that under "drift" conditions slow particles play an anomalously large role, so that for some systems the width of the nonlinear resonance retains the collision value (Γ) and drift broadening exerts almost no influence. However, in the systems normally used (absorption from the ground state of molecules) and in the standard equipment designs the influence of drift broadening is still large and various methods have been used or proposed to reduce it: increasing the light-beam diameter by means of telescope systems,[2,3] using spatially separated light beams,[6] or a molecular beam collinear with the light beam.[7] All of these measures involve serious technical difficulties and, moreover, do not result in total exclusion of drift broadening.

We shall now turn our attention to the following fact. Historically, it has turned out that most of the attention in theoretical analyses has been focused on low-order nonlinear resonance with respect to radiation intensity. This resonance gives the principal contribution when standard recording methods are used under conditions of slight field broadening. It turns out, however, that there is no drift broadening for higher-order nonlinear resonances because of the effect of slow particles.

We recall[5] that the slow-particle effect is caused by the inertia of the atomic oscillator: fast particles fly through the light beam without having time to "oscillate" and to interact with the field much less strongly than slow particles. It is obvious that the greater the inertia of interaction, the greater the degree of nonlinearity. This leads to a decrease of the effective velocity interval of the particles that contribute to the corresponding nonlinear resonance. In fact, even for a third-order nonlinear resonance with respect to the intensity of the standing wave the effective interval of "transverse" velocities is $\sim\Gamma a$ (a is the light-beam radius), which corresponds to a resonance width of $\sim\Gamma$.

We obtain an analytical expression for a third-order resonance. We assume that the particles interact with a monochromatic standing wave of frequency ω. We also assume that the intensity distribution is uniform in the light beam of radius a. For the probability of absorption $p(v, u; x, y)$ by particles with longitudinal (v) and transverse (u) velocities at a point with the transverse coordinates x and y, we obtain the following expression[8]:

$$p(v, u; x, y) = \frac{1}{2\pi i}\int_{\lambda - i\infty}^{\lambda + i\infty} e^{s(x+x_0)}\,p(v, u; s, y)\,ds, \quad x_0 = \sqrt{a^2 - y^2};$$

$$p(v, u; s, y) = \frac{|G|^2}{4} W(v, u)\,\frac{Y(us)}{s}\left\{1 + \frac{|G|^2}{4} Y(us)\left[\frac{1}{\Gamma_m + us} + \frac{1}{\Gamma_n + us}\right]\right\}^{-1},$$

$$Y(us) = 2(\Gamma + us)\left[\frac{1}{(\Gamma + us)^2 + (\Omega - kv)^2} + \frac{1}{(\Gamma + us)^2 + (\Omega + kv)^2}\right], \tag{1}$$

$$G = Ed_{mn}/2\hbar, \quad W(v, u) = \frac{2u}{\sqrt{\pi}\,\bar{v}^3} \exp\left[-(v^2 + u^2)/\bar{v}^2\right].$$

Here we have used a Laplace transform with respect to the variable x; the x axis is directed along the transverse velocity, E is the amplitude of the electric field of the wave, d_{mn} is a matrix element of the dipole moment, and Γ_m and Γ_n are the relaxation constants of levels m and n.

The increased role of slow particles (the appearance of additional powers of the combination us in the denominator), as we go from lower to higher orders in the expansion in powers of $|G|^2$, is easily traced in the expression $p(v, u; x, y)$. We calculate the absorption probability averaged over the velocities and over the transverse cross section of the beam

$$p = \frac{1}{\pi a^2} \int_{-\infty}^{\infty} dv \int_{0}^{\infty} du \int_{-a}^{a} dy \int_{-x_0}^{x_0} dx \, p(v, u; x, y). \tag{2}$$

Assuming, for simplicity, that $\Gamma = \Gamma_m = \Gamma_n$, we find that in the essentially "drift" situation ($\Gamma\bar{\tau} \equiv \Gamma a/\bar{v} \ll 1$)

$$p = \frac{|G|^2\sqrt{\pi}}{k\bar{v}} e^{-(\Omega/k\bar{v})^2}\left[1 - \beta(\Omega)\kappa + \gamma(\Omega)\kappa^2\right], \quad \kappa = |G|^2/\Gamma^2,$$

$$\beta(\Omega) = \frac{1}{2}(\Gamma\bar{\tau})^2\left[\ln\frac{1}{\Gamma\bar{\tau}} + \ln\frac{1}{\bar{\tau}\sqrt{\Gamma^2 + \Omega^2}}\right], \tag{3}$$

$$\gamma(\Omega) = \frac{3}{16}(\Gamma\bar{\tau})^2\left[\frac{1}{3} + \frac{\Gamma^2}{\Omega^2}\ln\left(1 + \frac{\Omega^2}{\Gamma^2}\right)\right].$$

The quantity κ is called the saturation parameter. Equation (3) for p in the absence of a term proportional to $\gamma(\Omega)$ was obtained previously in Ref. 5. The half-width δ_1 of the nonlinear resonance, which is described by the function $\beta(\Omega)$, is $\delta_1 = \sqrt{\Gamma/\bar{\tau}}$, i.e., it is still quite sensitive to the average transit time ($\bar{\tau}$) of the beam. The nonlinear resonance of the next order, described by the function $\gamma(\Omega)$, has a half-width $\delta_2 \cong 1.3\,\Gamma$, which is completely determined by collision broadening, consistent with the qualitative arguments presented above. The widths of nonlinear resonances of even higher orders are presumably determined by the value of Γ.

We have shown that the higher-order nonlinear resonances are not subject to a drift broadening. The possibility of detecting such resonances experimentally must therefore be considered. Higher-order nonlinear resonances have already been detected in an external absorbing cell.[9] Higher-order resonances can also be easily isolated in a laser with an internal nonlinear absorbing cell. Steady-state lasing can be represented as follows:

$$\tilde{\alpha}|G|^2[1-\tilde{\beta}(\Omega)\kappa-\gamma(\Omega)\kappa^2]=R|G|^2. \tag{4}$$

Here $\tilde{\alpha}$ is the gain which takes into account the losses in the absorbing cell, R is the loss factor associated with the escape of radiation from the cavity, $\tilde{\beta}(\Omega)$ includes the characteristics of the amplifying and absorbing media, and $\gamma(\Omega)$ is determined only by the absorbing cell. For a small saturation parameter it follows from Eq. (4) that

$$\kappa=\frac{\tilde{\alpha}-R}{\tilde{\alpha}\tilde{\beta}(\Omega)}-\frac{\gamma(\Omega)}{\tilde{\beta}(\Omega)}\left[\frac{\tilde{\alpha}-R}{\tilde{\alpha}\tilde{\beta}(\Omega)}\right]^2, \tag{5}$$

where κ characterizes the lasing power. For sinusoidal modulation of the loss factor R the signal at the second harmonic in the lasing power is proportional to the quantity $\gamma(\Omega)$. Thus a lower-order nonlinear resonance can be isolated by recording the second-harmonic signal. The described method is one way of isolating experimentally the higher-order nonlinear resonances, i.e., one way of obtaining a resonance not subject to a drift broadening.

1. V. S. Letokhov and V. P. Chebotaev, Printsipy nelineǐnoi lazernoǐ spektroskopii (Principles of Nonlinear Laser Spectroscopy), Nauka, Moscow, 1975.
2. C. Borde and J. L. Hall, Phys. Rev. Lett. **30**, 1101 (1973).
3. S. N. Bagaev, L. S. Vasilenko, V. G. Gol'dort, A. K. Dmitriev, and A. S. Dychkov, Kvantovaya Elektron. **4**, 1163 (1977) [Sov. J. Quantum Electron. **7**, 665 (1977)].
4. S. G. Rautian and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 686 (1969) [JETP Lett. **9**, 427 (1969)].
5. S. G. Rautian and A. M. Shalagin, Zh. Eksp. Teor. Fiz. **58**, 962 (1970) [Sov. Phys.–JETP **31**, 518 (1970)].
6. V. S. Letokhov and B. D. Pavlik, Opt. Spektrosk. **32**, 856 (1972) [Opt. Spectrosc. **32**, 455 (1972)]; V. P. Chebotayev, Appl. Phys. **15**, 219 (1978).
7. S. N. Bagaev and V. P. Chebotaev, Piz'ma Zh. Eksp. Teor. Fiz. **16**, 614 (1972) [JETP Lett. **16**, 433 (1972)].
8. A. M. Shalagin, Candidate's dissertation, Novosibirsk, Inst. Nucl. Phys., Siberian Branch, USSR Acad. Sci., 1973.
9. S. G. Rautian, E. G. Saprykin, V. A. Sorokin, and A. M. Shalagin, Kvantovaya Elektron. **7**, 1354 (1980) [Sov. J. Quantum Electron. **10**, 779 (1980)].

Translated by Eugene R. Heath
Edited by S. J. Amoretty