Eur. Phys. J. D **1**, 217–221 (1998)

THE EUROPEAN PHYSICAL JOURNAL D

Nonlinear diffraction due to the transient polarization in a thin film of atomic gases*

T.A. Vartanyan[a] and D.L. Lin[b]

Department of Physics, State University of New York at Buffalo, Buffalo, New York 14260-1500, USA

Received: 3 July 1997/Accepted: 5 December 1997

Abstract. Nonlinear optical response of a thin layer of rarefied atomic vapor is examined by taking into account the atomic motion as well as collisions with the cell walls. Extraordinary pattern of self-diffraction spectrum due to the transient polarization is predicted. It is shown that the spectra are Doppler free and depend strongly upon the vapor thickness. A new possibility of signal enhancement is revealed and a simple device is suggested to enhance greatly the nonlinear reflection signal from a resonant vapor layer.

PACS. 32.70.Jz: Line shapes, widths, and shifts – 42.40.Lx: Diffraction efficiency, resolution, and other hologram characteristics – 42.25.Gy: Edge and boundary effects, reflection and refraction

In spite of the great progress in the fabrication of such novel nonlinear optical materials as photo-refractive crystals and quantum well structures in recent years, a simple glass cell containing a resonant atomic vapor is still considered as a promising nonlinear device. Dynamic holography, optical phase conjugation and real-time signal processing are among the possible applications of atomic vapors as a nonlinear medium [1]. Atomic vapors have such a large and fast nonlinear response on or near resonances that they are capable of processing signals and images with milliwatt lasers at submicrosecond time scales. Moreover, the physical mechanism that leads to the optical nonlinearity seems to be perfectly understood, and can be well adapted for the physical reinterpretation of experiments [2].

Although rarefied atomic vapor is the simplest possible nonlinear optical medium, conventional dispersion theory fails to describe selective reflection on its boundary even in the linear domain. When the Doppler width becomes larger than the homogeneous line width, the mean free path of the atom becomes larger than the wavelength. Consequently, the atomic response of the vapor on the external field becomes nonlocal. The atomic motion, and the collision between atoms and the solid wall in particular, must be explicitly included in the treatment, or the most important feature of the reflection spectrum will be lost. The most prominent manifestation of this phenomenon is found in the selective reflection [3–8] at the interface of a transparent dielectric solid and atomic vapor.

When the incident light is in resonance with atoms leaving the interface, the selective reflection is substantially reduced for positive detunings. This reduction is a consequence of surface quenching of atomic polarization. On the other hand, for negative detunings, when light is in resonance with atoms flying towards the surface, one finds an extra contribution that is absolutely unexpected from atoms leaving the surface. This is due to the transient polarization induced by atom-wall collisions. This contribution is identical to that of atoms flying in opposite directions with the same speed. Hence the overall velocity average is twice the average over arriving atoms alone. The discontinuity at zero velocity leads to a sharp feature in selective reflection spectrum.

There has been a great deal of interest in recent years in the transient effect on nonlinear properties of reflection from a single solid-vapor interface [9–18]. The purpose of this letter is to report the unexpected diffraction pattern from a thin layer of atomic vapor sandwiched between transparent dielectric solids from our study of the interplay of transient and interference effects. In the linear regime, novel phenomena found in selective reflection from such systems have recently been reported [19].

Consider two laser beams of amplitudes E_1, E_2 and wave vectors (k_{1x}, k_{1z}), (k_{2x}, k_{2z}) but the same frequency ω at the near-normal incidence on a boundary interface of a vapor layer. The layer thickness can be anywhere between the wavelength and the optical penetration depth, which is defined as the inverse of the absorption coefficient of the vapor. This range of thickness is large enough because the Doppler broadening leads to reduction of the absorption coefficient. Hence the penetration depth is

* Supported by a grant from National Academy of Science/National Research Council Cooperation in Applied Science and Technology program

[a] *Permanent address*: State Optical Institute, Divenskaya 18-17, St. Petersburg 197061, P.O. Box 656, Russia

[b] e-mail: dllin@acsu.buffalo.edu

always far larger than the wavelength. More specific conditions of validity for nonlinear diffraction will be worked out explicitly in the following.

For simplicity, we assume only two-level atoms with a transition frequency ω_0 in the vapor. We exclude the large-angle incidence in which case the x-component of atomic motion leads to large Doppler broadening of all resonance features in the frequency range. Moreover, the induced grating of nonlinear refraction index will be washed out because the mean free path of an excited atom is much larger than the wavelength while the fringe spacing may be only a half of wavelength. As a consequence, the nonlinear response of the vapor diminishes.

For nearly normal incidence, we have $k_{1x},\ k_{2x} \ll k$, $k_{1z},\ k_{2z} \approx k$ where $k = \omega/c = \sqrt{k_{ix}^2 + k_{iz}^2}$. Then the sum of two beams $E_1 \exp(ikz + ik_{1x}x)$ and $E_2 \exp(ikz + ik_{2x}x)$ at nearly normal incidence can be regarded as a single beam $E(x)\exp(ikz)$ with slowly varying amplitude

$$E(x) = E_1 \exp(ik_1 x) + E_2 \exp(ik_2 x). \tag{1}$$

As the amplitude is nearly constant, its derivative with respect to x is neglected in the following calculations. Thus the ordinary third order nonlinear term $E(x)|E(x)|^2$ lead to small corrections to both the specularly reflected and self-diffracted beams which propagate in different directions. The reflected beams have wave vectors $(k_{1x} - k)$ and $(k_{2x} - k)$, and the self-diffracted waves are given by

$$E_3(x) \sim E_2^2 E_1^* \exp[i(2k_{2x} - k_{1x})x], \tag{2a}$$

$$E_4(x) \sim E_1^2 E_2^* \exp[i(2k_{1x} - k_{2x})x], \tag{2b}$$

with the corresponding wave vectors $(2k_{2x} - k_{1x},\ -k)$ and $(2k_{1x} - k_{2x},\ -k)$. A schematic sketch of the situation is shown in Figure 1. These particular beams that can not be produced on the boundary of linear medium are of special interest in applications. For $k_{1x} = 0$, the beam E_1 is exactly at the normal incidence. In this particular case, E_4 represents the amplitude of the beam that is phase conjugated to E_2.

The diffraction efficiency is determined by the solution of Bloch equation [20] for the resonant atom. In the two-level approximation, this is a set of first order differential equations for the off- diagonal matrix element ρ and the population difference n. For a particular group of atoms with $v_z = v$, one can under the steady state obtain the following set of equations

$$v\frac{\mathrm{d}\rho}{\mathrm{d}z} + (\gamma_2 - i\Delta)\rho = \frac{iDE}{2\hbar}\mathrm{e}^{ikz}n, \tag{3a}$$

$$v\frac{\mathrm{d}n}{\mathrm{d}z} + \gamma_1 n = \gamma_1 + \frac{iDE^*}{\hbar}\mathrm{e}^{-ikz}\rho - \frac{iDE}{\hbar}\mathrm{e}^{ikz}\rho^* \tag{3b}$$

where D denotes the atomic dipole moment, $\Delta = \omega - \omega_0$ stands for the detuning, and γ_1 and γ_2 are relaxation constants for the population (diagonal elements) and coherence (off-diagonal elements) of the density matrix, respectively. The boundary conditions are chosen such that all atoms are unexcited when they leave the surfaces, namely,

$$\rho(z = 0, v > 0) = \rho(z = l, v < 0) = 0, \tag{4a}$$

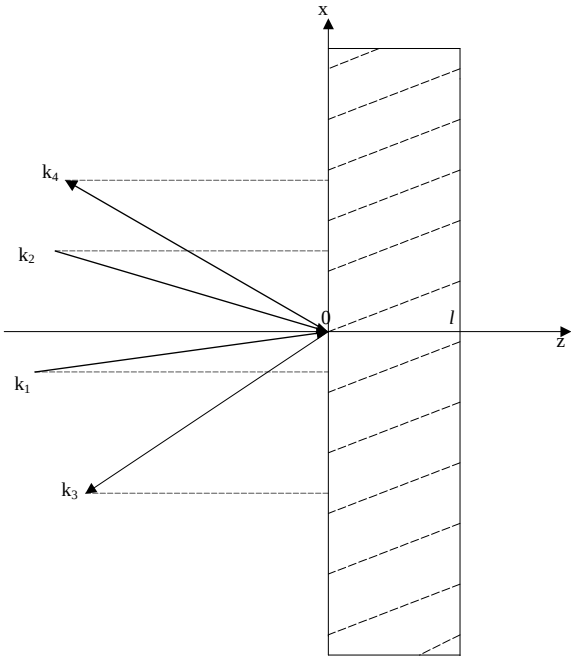

Fig. 1. Schematic sketch of the geometry. k_1, k_2 represent the incident beams and k_3, k_4 indicate the self-diffracted waves.

$$n(z = 0, v > 0) = n(z = l, v < 0) = 1. \tag{4b}$$

In the limit of large Doppler broadening, the optical density of the atomic vapor may be so small that we can apply the perturbation theory. We start with the incident field as a driving field in E_3 and E_4, which are solved with boundary conditions (4). From the density matrix thus obtained, we can find the polarization from

$$P(z) = 2ND\langle\rho(z, v)\rangle \tag{5}$$

where N is the density of atoms and $\langle\ \rangle$ means the average over Maxwellian distribution of all velocities. The reflected field is given by

$$E_\mathrm{r} = 2\pi ik \int_0^l P(z)\mathrm{e}^{ikz}\mathrm{d}z \tag{6}$$

where $E_\mathrm{r} = E_\mathrm{r}^> + E_\mathrm{r}^<$. $E_\mathrm{r}^>$ is the field due to atoms leaving the surface at $z = 0$ with $v > 0$, and $E_\mathrm{r}^<$ due to atoms leaving the surface at $z = l$ with $v < 0$.

To calculate these components explicitly, one has to solve for any fixed x the set of equations (3) for atoms with specified v. We restrict, for simplicity, our discussion to the particular case of equal relaxation constants $\gamma_1 = \gamma_2 = \gamma$. From the Laplace transformation of the population function $n(z)$ and density matrix $\rho(z)$

$$\hat{n}(p) = \int_0^\infty \mathrm{e}^{-pz}n(z)\mathrm{d}z, \tag{7a}$$

$$\hat{\rho}^>(p) = \int_0^\infty \mathrm{e}^{-pz}\rho^>(z)\mathrm{d}z. \tag{7b}$$

We can transform the set of equations (3) with boundary conditions (4) into a set of algebraic equations and find its solution in the form

$$\hat{\rho}(p) = \frac{iDE}{2\hbar}\Bigg\{(\gamma - i\Lambda + vp)(p - ik) \times\left[1 + \frac{D^2E^2/\hbar^2}{(\gamma - i\Lambda + vp)(\gamma + i\Delta - 2ikv + vp)}\right]\Bigg\}^{-1}. \quad (8)$$

In the case of a thick vapor layer, $l \to \infty$, equation (6) has the same form as equation (7b) if one identifies $p = -ik$. Hence $E_{\rm r}^{>}$ can be expressed in terms of $\hat{\rho}^{>}(-ik)$. For a thin vapor layer we are dealing with here, l remains finite and we have to use the Laplace inversion formula

$$\rho^{>}(z) = \frac{1}{2\pi i}\int_{q-i\infty}^{q+i\infty}\hat{\rho}^{>}(p)\mathrm{e}^{pz}\mathrm{d}p \quad (9)$$

to find the polarization $P^{>}(z)$ according to equation (5). In equation (9), the positive number q is so chosen that all the poles p_n of $\hat{\rho}(p)$ are found at the left of q, namely, $\mathrm{Re}(p_n) < q$. The integral in equation (9) can be evaluated by closing the path in the left half of the complex p plane with the result

$$E_{\rm r}^{>} = 4\pi ikND \times\left\langle\rho^{>}(-ik) + \sum_{n=1}^{3}\frac{\exp[(p_n + ik)l]}{p_n + ik}\operatorname*{res}_{p_n}[\rho^{>}(p)]\right\rangle_{v>0} \quad (10)$$

where $\mathrm{res}_{p_n}[\rho(p)]$ stands for the residue of the function $\hat{\rho}(p)$ at the zeros $p = p_n$ of its denominator.

In what follows, we introduce for convenience the dimensionless variables $\phi = kl$, $y = v/v_{\rm T}$, $\Omega = \Delta/kv_{\rm T}$, $\Gamma = \gamma/kv_{\rm T}$ and $\epsilon = DE/\hbar kv_{\rm T}$, where $v_{\rm T}$ is the most probable thermal velocity. With this notation, one finds after a straightforward but tedious calculation

$$E_{\rm r}^{>} = \sqrt{\pi}ND\varepsilon\left\{I_1 + [I_2 + I_3 + I_4]\mathrm{e}^{2i\phi}\right\} \quad (11)$$

$$I_1 = \int_0^\infty \frac{\exp(-y^2)\mathrm{d}y}{(y + \Omega + i\Gamma)\left[1 - \frac{|\varepsilon|^2}{(i\Gamma + \Omega + y)(3y - \Omega + i\Gamma)}\right]} \quad (11a)$$

$$I_2 = \int_0^\infty \frac{\exp(-y^2)\mathrm{d}y}{(y - \Omega - i\Gamma)[1 + \frac{|\varepsilon|^2}{(y - \Omega - i\Gamma)(y - \Omega + i\Gamma)}]} \quad (11b)$$

$$I_{3,4} = -\int_0^\infty \frac{y\exp[-y^2 + (-\Gamma \pm iR)\phi/y][1 \pm (\Omega - y)/R]\mathrm{d}y}{[2y \pm R + i\Gamma][\pm R + i\Gamma]}. \quad (11c)$$

where "+" in "±" refers to I_3 and "−" refers to I_4, and we have defined $R = \sqrt{|\varepsilon|^2 + (\Omega - y)^2}$.

The saturation intensity for a resonant transition in a rarefied atomic vapor lies in the range of 10 mW/cm^2, and is readily achievable by modern CW lasers. We are, however, interested in the case of relatively low intensity in which the integrals I_1 and I_2 may be expanded into powers of the parameter $|\varepsilon|^2/\Gamma^2 \ll 1$. This means that we restrict our discussions to the case when the two-level system is far from saturation. Similar expansion is not valid for I_3 and I_4 because of the singularity at $y = \Omega$. Despite of this artificial singularity, it can be shown that for $\phi \gg 1$, the nonlinear contribution to I_3 and I_4 is generally smaller than I_1 and I_2 by a factor of ϕ, and hence can be safely ignored in the calculation provided that conditions describe above are satisfied. This is in contrast to the linear effect [19] in which all four integrals are of comparable size.

The reflected field $E_{\rm r}^{<}$ from atoms moving in the opposite direction can be written down directly from equation (10) by means of the following procedure: change the sign of v, replace E by $E\exp(-ikl)$, take the complex conjugate of the resulting expression, change the overall sign and multiply the result by $\exp(ikx)$. The combined result then represents the nonlinear part of the reflection

$$E_{\rm r}^{\rm NL} = \sqrt{\pi}ND\varepsilon|\varepsilon|^2\Bigg\{\int_0^\infty\frac{\exp(-y)^2\mathrm{d}y}{(y + \Omega + i\Gamma)^2(3y - \Omega + i\Gamma)} - \int_0^\infty\frac{\exp(-y^2)\mathrm{d}y}{(y + \Omega + i\Gamma)^2(y + \Omega - i\Gamma)} + \mathrm{e}^{2i\phi}\Bigg(\int_0^\infty\frac{\exp(-y^2)\mathrm{d}y}{(y - \Omega - i\Gamma)^2(3y + \Omega - i\Gamma)} - \int_0^\infty\frac{\exp(-y^2)\mathrm{d}y}{(y - \Omega - i\Gamma)(y - \Omega + i\Gamma)}\Bigg)\Bigg\}. \quad (12)$$

It is noted that the amplitude depends on x through $\varepsilon|\varepsilon|^2 \sim E(x)|E(x)|^2$.

The diffraction spectrum is proportional to $|E_{\rm r}^{\rm NL}|^2$ which is plotted in Figure 2 for several choices of the vapor-layer thickness with $\gamma = 0.02$. For selective reflection, we are mainly concerned with the region around the resonance. In the limit of $\Omega \to 0$, all integrals can be evaluated analytically. The result looks more naturally when the detuning is normalized to the homogeneous width γ rather than the Doppler width $kv_{\rm T}$. Thus we have, in terms of $\nu = \Delta/\gamma$ instead of Ω,

$$E_{\rm r}^{\rm NL} = \frac{\sqrt{\pi}}{2}ND\varepsilon\frac{|\varepsilon|^2}{\Gamma^2}\Bigg\{(1 + \mathrm{e}^{2i\phi})\Bigg[\frac{2i\nu}{(1 - i\nu)(1 - 2i\nu)} - i\tan^{-1}\nu - \frac{3}{2}\frac{\ln 3 - 2i\tan^{-1}\nu}{(1 - 2i\nu)^2}\Bigg] + (1 - \mathrm{e}^{2i\phi})\frac{i\pi}{2}\Bigg\}. \quad (13)$$

The thickness dependence of the diffraction efficiency may be traced analytically at the resonance $\nu = 0$, that is,

$$E_{\rm r}^{\rm NL}(\nu = 0) = \frac{\sqrt{\pi}}{2}ND\varepsilon\frac{|\varepsilon|^2}{\Gamma^2}\mathrm{e}^{i\phi}[\pi\sin\phi - 3\ln 3\cos\phi]. \quad (14)$$

It is easily seen from equation (14) that the peak intensity $|E_{\rm r}^{\rm NL}(\nu = 0)|^2$ at resonance reaches its maximum when

$$\phi = (n + 1/2)\pi + \tan^{-1}(\pi/3\ln 3) \approx (n + 3/4)\pi \quad (15)$$

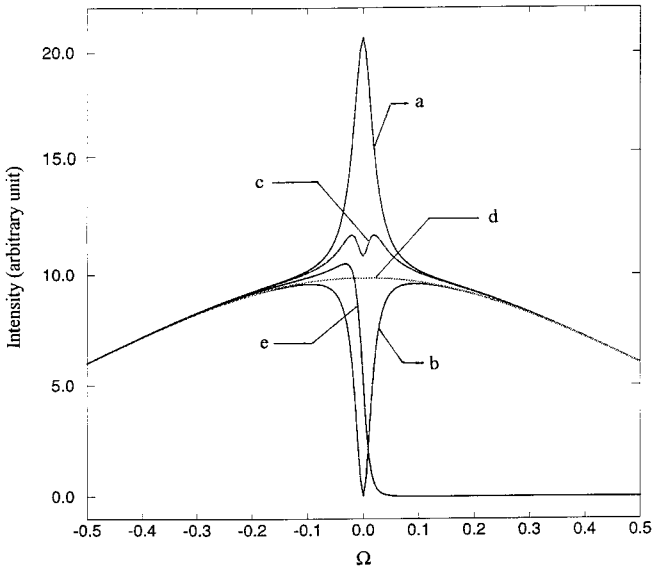

Fig. 2. Diffraction efficiency (in arbitrary unit) from a thin layer of resonant atomic vapor *versus* the dimensionless detuning for four choices of the vapor thickness: (a) $l = (n+3/4)\lambda/2$, (b) $l = (n + 1/4)\lambda/2$, (c) $l = n\lambda/2$ and (d) $l = (n + 1/2)\lambda/2$. The corresponding result for a thick vapor layer ($l \to \infty$) is also shown by (e). The integer n is arbitrarily chosen within the range $|\Omega/\pi\Gamma|$ and $|1/\pi\Gamma|$ and $\Gamma = 0.02$.

for positive integer n. This is curve (a) in Figure 2. Curve (b) shows no diffracted beam at the resonance when

$$\phi = n\pi + \tan^{-1}(3\ln 3/\pi) \approx (n + 1/4)\pi, \qquad (16)$$

which represents a sequence of nodes in the diffraction spectrum.

When $\phi = n\pi$, the second term in equation (13) vanishes and the spectrum is shown by curve (c). Since we are discussing the thickness dependence of the nonlinear reflection at $\nu = 0$, the spectrum has a period of $\lambda/2$. Hence it does not matter whether n is even or odd. For the special case of $\phi = (n + 1/2)\pi$, the first term in equation (13) vanishes and the spectrum is given by curve (d). This is just the conventional case when there is no transient effect. It is observed that the sub-Doppler structure disappears and the peak intensity in this case is only half of that in (a). For comparison purpose, the corresponding spectrum for a single interface is also shown as curve (e) which follows from equation (13) by deleting all terms involving $\exp(2i\phi)$.

There are a number of new features for this nonlinear diffraction spectrum due to the transient polarization. We have seen that the central peak intensity is enhanced by a factor of two in comparison with the single boundary or thick vapor case. Recall that the ordinary interference pattern at the diffraction center is determined by $|1 - e^{2i\phi}|^2$ with a sequence of nodes at $\phi = n\pi$. Equation (16), however, predicts that the nodes are shifted by an amount

$$\tan^{-1}(3\ln 3/\pi) \approx 0.809, \qquad (17)$$

and according to equation (15) the peaks are shifted from their ordinary positions at $\phi = (n + 1/2)\pi$ by

$$\tan^{-1}(\pi/3\ln 3) \approx 0.761. \qquad (18)$$

Novel interference effects discussed above are likely to play an important role in an extra thin ($\sim 10\ \mu$m) cesium vapor cell [21]. Cesium vapor is perhaps the best choice for observing such effects. Its large saturation pressure ensures that extreme cell temperatures may not be necessary. For example, at only 210 °C the concentration of cesium atoms can be as large as 4×10^{15} cm^{-3}. On the other hand, cesium resonant transition at $\lambda = 852$ nm may be excited with modern tunable CW lasers without difficulty.

Both the Ti:saphire laser [21] and diode laser [22] have been employed recently for this purpose. It is demonstrated that an intensity of 3 W/cm^2 or higher can readily be achieved by focusing the laser beam to a spot of about 1 mm in diameter. A vapor wedge can be created for the study of the thickness dependence of nonlinear selective reflection by two nonparallel surfaces of transparent solid. One side of the surfaces is in contact with each other and the other side is separated by a thin gasket. As an estimate for practical purposes, we take the dipole moment of resonant transition $D = 1.9\times10^{-29}$ °Cm, Doppler width $kv_{\rm T} = 270$ MHz and the self-broadening constant 1.5×10^{-7} Hz/cm^3 [11,14]. Then we find, according to equation (14), the ratio of the diffracted beam intensity to the incident intensity to be $|E_{\rm r}^{\rm NL}/E|^2 \approx 10^{-4}$, which is large enough not only for observing the effect but also for some applications as well.

In conclusion, we remark that the present investigation opens a new possibility to enhance the nonlinear reflection signal from a resonant vapor layer. Consider a sequence of N parallel plates of transparent material inserted inside the vapor cell. The optical thickness of a plate plus the interplate distance is the optical distance L between two corresponding interfaces. If l is so chosen that equation (15) is satisfied and L is a multiple of half-wavelength, all the nonlinear contributions will interfere constructively. As a consequence, the nonlinear signal may be enhanced by a factor up to N^2. This means that the peak intensity can be as strong as $2N^2$ times that from a thick vapor. The only point one has to notice is that the thickness l of every vapor layer in the cell is small compared to the absorption length. This research was supported by a grant from National Academy of Science/National Research Council Cooperation of Applied Science and Technology program, and by the Russian Federation State Program "Basic Metrology".

References

1. See, for example, B. Ai Glassner, R.J. Knize, J.P. Partenan, R.W. Hellwarth, in *International Quantum Electronics Conference*, **9**, 1994 OSA Technical Digest Series (Optical Society of America, Washington, D. C. 1944).
2. J.J. Maki, W.V. Davis, R.W. Boyd, J.E. Sipe, Phys. Rev. A **46**, 7155 (1992).

3. J.L. Cojan, Ann. Phys. France **9**, 358 (1954).
4. M.F.J. Schuurmans, J. Phys. France **37**, 469 (1976).
5. A.M. Akushin, V.L. Velichanskii, A.S. Zibrov, V.V. Sautenkov, E.K. Yurkin, N.V. Senkov, JEPT Lett. **36**, 303 (1982).
6. M. Oria, M. Chevrollier, D. Bloch, M. Fichet, M. Ducloy, Europhys. Lett. **14**, 527 (1991).
7. J. Guo, J. Cooper, A. Gallagher, M. Lewenstein, Opt. Commun. **110**, 197 (1994).
8. P. Wang, A. Gallagher, J. Cooper, Phys. Rev. A **56**, 1598 (1997).
9. T.A. Vartanyan, Sov. Phys. JETP **61**, 674 (1985).
10. T.A. Vartanyan, Opt. Spectrosc. (USSR) **68**, 365 (1990).
11. A.M. Akushin, V.L. Velichanskii, T.A. Vartanyan, R.G. Gamidov, V.A. Sautenkov, S.I. Filimonov, Opt. Spectrosc. (USSR) **66**, 423 (1989).
12. S. Singh, G.S. Agarwal, Opt. Communic. **59**, 107 (1986).
13. G. Nienhuis, F. Schuller, M. Ducloy, Phys. Rev. A **38**, 5197 (1988).
14. A.M. Akulshin, A.A. Celikov, V.A. Sautenkov, T.A. Vartanyan, V.L. Velichansky, Opt. Communic. **85**, 21 (1991).
15. A.E. Korolyev, V.N. Nazarov, D.I. Staselko, Sov. Tech. Phys. Lett. **15**, 824 (1989).
16. F. Schuller, G. Niehuis, M. Ducloy, Phys. Rev. A **43**, 443 (1991).
17. V.A. Sautenkov, R.G. Gamidov, A. Weis, Phys. Rev. A **55**, 3137 (1997).
18. B. Gross, N. Papageorgiou, V. Sautenkov, A. Weis, Phys. Rev. A **55**, 2973 (1997).
19. T.A. Vartanyan, D.L. Lin, Phys. Rev. A **51**, 1959 (1995).
20. L. Allen, J.H. Eberly, *Optical Resonance and Two-Level Atoms* (John-Wiley, New York, 1975).
21. D.S. Glassner, B. Ai, R.J. Knize, Optics Lett. **19**, 207 (1994).
22. O.A. Rabi, A. Amy-Klein, S. Saltei, M. Ducloy, Europhys. Lett. **25**, 579 (1994).