

# Effects of Quantum Statistics in Polarization-Sensitive Spectroscopy of Free and Interacting Atoms

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Received July 3, 2001

**Abstract**—Spectroscopic applications of radiation with nonclassical statistics are discussed. Applications where the second-order correlation function of the field intensity is a measurable quantity are considered. It is shown that the spectroscopy of intensity fluctuations allows the spectral content of light scattered into modes of squeezed radiation by a small number of scatterers to be measured with a higher efficiency than in the case of scattering into vacuum modes. Another example considered in this paper is related to two-photon polarization-dependent excitation of an atom colliding with another atom in the case when photons initiating this transition are in a cooperative polarization-entangled state. It is shown that a specific polarization dependence of two-photon absorption probability arising in this case cannot be interpreted in terms of classical concepts. This dependence allows the internal dynamics of a collisional process to be controlled to some extent.

## 1. INTRODUCTION

Analysis of possible spectroscopic applications of light with nonclassical statistics (in a squeezed or entangled state) is a promising, but not very well-developed direction of modern quantum optics. There are not so many papers directly dealing with this problem [1–13]. Of those, only two works are experimental [6, 7]. The first experimental paper demonstrates the improvement of the threshold sensitivity in absorption spectroscopy related to the squeezing effect. The second experiment reveals the linear dependence of the two-photon absorption probability on the intensity of pump light, which was earlier predicted in several theoretical papers [1, 4, 5]. We can easily understand why the number of papers devoted to this problem is small if we take into consideration that any experiment with nonclassical light is rather difficult to implement and should be, therefore, very well motivated. Spectroscopic applications is not an exception. Indeed, the advantages of nonclassical light in spectroscopic problems are often understood as the improvement of the signal-to-noise ratio in measurements [6], while the advantages of nonclassical light for studying elementary objects are much less obvious.

In this paper, we will consider two examples that provide, in our opinion, a motivation for such experiments. In the first part of this paper, we will consider the well-known and thoroughly tested method of intensity fluctuation spectroscopy (IFS) [14, 15]. This technique is, in fact, one of the modifications of high-resolution spectroscopy, which allows the spectra of Rayleigh and Raman scattering of optical radiation to be observed in those cases when frequency differences and the spectral distributions themselves lie within the radio-frequency range. We will demonstrate that the use of nonclassical radiation permits the observation of

such spectra in the case of a small number of scatterers or even a single scatterer.

Another example considered in this paper is related to two-photon polarization-dependent excitation of an atom colliding with another atom. In this analysis, we will assume that the photons initiating this transition are in a cooperative polarization-entangled state, i.e., they are being created through spontaneous parametric scattering of the second type. We will show that this process is characterized by a specific polarization dependence of the two-photon absorption probability, which cannot be interpreted in terms of classical concepts and which allows the internal dynamics of a collisional process to be controlled to a certain extent.

A fundamental common feature of the two examples considered in this paper is that the observed effects are determined in both cases by nonclassical properties of the second-order correlation function of light intensity.

## 2. SPECTROSCOPY OF INTENSITY FLUCTUATIONS WITH THE USE OF SQUEEZED LIGHT

The IFS method is based on the idea of using the informative content of fluctuations of radiation parameters for analyzing the state of a medium under study. Measurement of the second-order intensity correlation function opens an additional and, to a certain extent, independent channel of extracting the information concerning the medium. The IFS method is, in fact, one of the modifications of high-resolution spectroscopy exploiting the fact that the interaction with a medium gives rise to deviations of the statistics of interrogating and scattered radiation from Gaussian and Poissonian statistics. In this paper, we are interested in the case when probe radiation initially possesses nonclassical

statistical properties, residing in a squeezed state. In such a situation, we can expect, as shown in [11], that the sensitivity of the IFS method can be substantially improved. However, because of specific properties of squeezed radiation, the scheme of measurements requires an additional discussion.

In this paper, we will restrict ourselves to the discussion of how the contributions related to Raman or Rayleigh scattering can be detected and selected in such a more sophisticated modification of the IFS method. Raman and Rayleigh scattering can be observed in any direction. Assuming that the signal is detected at an arbitrary angle excluding the case of forward scattering, we ignore effects related to a parametric interaction. We will show that some interesting generalizations can be expected for the IFS method based on the principle of optical heterodyning with detection of radiation scattered not to a vacuum, but to modes preliminarily filled with squeezed radiation. These generalizations may be expected, first of all, for the case of a small number of scatterers. In our analysis, we will use general concepts of the nature of scattering particles without discussing the details of the elementary scattering event itself. We believe that the results of our analysis may serve as a theoretical background for a new modification of the IFS method as an experimental technique employing the quantum properties of interrogating radiation.

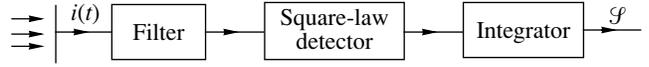
The correlation function of the photocurrent  $G(\tau) = \langle i(\tau)i(0) \rangle$  is an observable quantity in the IFS method. In the case of a stationary random process  $i(t)$ , this function can be represented as the following expansion:

$$G(\tau) = \bar{i}^2 + \bar{i}\delta(\tau) + G_{\text{inf}}(\tau). \quad (1)$$

The spectral density of a random process is determined by the relevant Fourier expansion:

$$\begin{aligned} G(\Omega) &= \int_{-\infty}^{\infty} G(\tau)e^{i\Omega\tau}d\tau = \bar{i}^2 2\pi\delta(\Omega) + \bar{i} + G_{\text{inf}}(\Omega) \\ &= \bar{i}^2 2\pi\delta(\Omega) + \bar{i}(1 + \zeta\xi(\Omega)). \end{aligned} \quad (2)$$

Here,  $\bar{i} = \zeta JS_0$  is the mean photocurrent,  $\zeta$  is the quantum efficiency of the photodetector,  $J$  is the photon fluence, and  $S_0$  is the illuminated surface area of the photocathode. With a heterodyning detection scheme in mind, we assume that  $S_0$  coincides with the cross section of the reference wave and  $J$  is determined by the photon fluence in the reference wave. The first two terms on the right-hand sides of Eqs. (1) and (2) correspond to the uncorrelated and shot-noise components of the correlation function and the photocurrent spectrum. The last term describes the so-called informative contribution, which is determined by the spectral content of scattered radiation. The second equality in Eq. (2) relates the informative part  $G_{\text{inf}}(\Omega)$  to the spectral Mandel parameter  $\xi(\Omega)$ , which characterizes the



**Fig. 1.** Detection channel. The current from the output of the photodetector passes through a resonant filter tuned to the frequency  $\tilde{\Omega}$ , undergoing quadratic detection and averaging with an integrator.

relative deviation of the photocurrent spectrum from the Poissonian level.

Figure 1 shows one of the modifications of the detection channel that can be used to measure the photocurrent spectrum. When the photocurrent spectrum is analyzed with the use of a narrow-band filter with a bandwidth  $\gamma_f$ , characterized by the spectral transfer function  $Y(\Omega)$  and the signal accumulation time  $T$ , the detected output signal  $\mathcal{S}$  is proportional to the spectral density  $G(\Omega)$  of a random process at the filter frequency  $\tilde{\Omega}$ :

$$\begin{aligned} \mathcal{S} &= \mathcal{S}(\tilde{\Omega}) \\ &= \int_0^{\infty} G(\Omega)|Y(\Omega)|^2 \frac{d\Omega}{\pi} \approx G(\tilde{\Omega}) \int_0^{\infty} |Y(\Omega)|^2 \frac{d\Omega}{\pi}. \end{aligned} \quad (3)$$

For a stationary process, the mean value of  $\mathcal{S}$  depends neither on the choice of the initial moment of time nor on the signal averaging time  $T$ .

The second equality in Eq. (2) defines the spectral Mandel parameter  $\xi(\Omega)$ , which characterizes the deviation of the fluctuation level from the shot-noise limit in the considered spectral range. The spectral dependence of the Mandel parameter is determined by the relevant dependences of field correlation functions and is due to quantum beats of the components of the scattered wave with the reference wave. It follows directly from Eq. (3) that the measurement of the observable  $\mathcal{S} = \mathcal{S}(\tilde{\Omega})$  corresponds to the measurement of the Mandel parameter  $\xi(\tilde{\Omega})$  in the vicinity of the resonant filter frequency  $\Omega \approx \tilde{\Omega}$ :

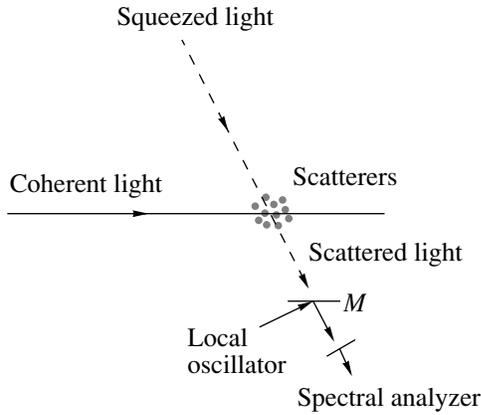
$$\mathcal{S} = \mathcal{S}(\tilde{\Omega}) = \eta_0 \gamma_f \bar{i} (1 + \zeta \xi(\tilde{\Omega})), \quad (4)$$

where

$$\eta_0 = \frac{1}{\gamma_f} \int_0^{\infty} |Y(\Omega)|^2 \frac{d\Omega}{\pi} \sim O(1). \quad (5)$$

Here, we took into consideration that the uncorrelated contribution in the spectrum (2) is suppressed by the filter and never appears in the output signal.

The problem of estimating the variance of the quantity  $\mathcal{S} = \mathcal{S}(\tilde{\Omega})$  and, correspondingly, the accuracy of measurement of the informative part of the photocurrent spectrum contained by the spectral Mandel param-



**Fig. 2.** Measurement of Raman scattering spectra by the IFS method. An additional squeezed light beam propagates in the direction of observation. At the output of the scattering medium, the scattered and squeezed light beams are mixed with a reference wave on mirror  $M$ , whose transmission coefficient is close to unity and the reflection coefficient is low. Then, the signal undergoes spectral analysis, as shown in Fig. 1.

eter is much more complicated. We will use the results of the paper [11], which shows that, in the case when nonclassical squeezed radiation is incident upon the photodetector surface, the sought-for variance is also determined by the spectral Mandel parameter at the filter frequency:

$$\Delta \mathcal{G}^2 = 2\eta_1 \frac{\gamma_f T}{i} (1 + \zeta \xi(\tilde{\Omega}))^2, \quad (6)$$

where

$$\eta_1 = \frac{1}{\gamma_f} \int_0^\infty |Y(\Omega)|^4 \frac{d\Omega}{\pi} \sim O(1). \quad (7)$$

Relationship (6), which is valid in the limiting case of  $\gamma_f T \rightarrow \infty$ , can be employed to estimate the threshold sensitivity in measurements of the Mandel parameter by the method of optical heterodyning.

Let us represent the Mandel parameter as

$$\xi(\Omega) = \xi_0 + \delta\xi(\Omega), \quad (8)$$

where  $\xi_0$  is the constant value of this parameter corresponding to radiation reaching a photodetector either in the absence of a scattering medium or in the case when measurements are performed for those spectral ranges where no scattering occurs and  $\delta\xi = \delta\xi(\Omega)$  is a small correction determined by the interaction of interrogating radiation with the medium under study. Using Eq. (8), we arrive at the following estimate for the limiting value  $\delta\xi$  that can still be detected in experiments:

$$\delta\xi \approx \frac{2\sqrt{\eta_1}}{\zeta \eta_0} \frac{1}{(\gamma_f T)^{1/2}} (1 + \zeta \xi_0). \quad (9)$$

This estimate shows that the sensitivity of measurements of the informative component of the photocurrent spectral density can be, in principle, improved relative to the IFS scheme based on scattering to vacuum modes provided that radiation is scattered to the modes filled with squeezed radiation with the unperturbed Mandel parameter  $\xi_0 \rightarrow -1$  and the photodetector efficiency  $\zeta \rightarrow 1$ .

Let us consider for the sake of definiteness IFS applications for studying Raman scattering. The case of Rayleigh scattering can be analyzed in a similar way. Figure 2 shows the scheme of detection. Importantly, squeezed radiation comes in the direction of detection of scattered radiation. Detection is performed at an arbitrary angle except for the regime of forward scattering. Since phase matching is not achieved for radiation propagating in the lateral direction, the modes of squeezed radiation do not interact with the classical wave. This implies that the modes of squeezed radiation may change only due to linear absorption and dispersion. The change in the spectral Mandel parameter due to Raman scattering combined with the effect of linear absorption is given by

$$\delta\xi(\Omega) = -(\kappa_1 + \kappa_2)\xi_0(\Omega) + \Lambda_1(\Omega) + \Lambda_2(\Omega), \quad (10)$$

where  $\kappa_1$  and  $\kappa_2$  are the linear absorption coefficients at the Raman frequencies  $\omega_1 = \omega + \Omega$  and  $\omega_2 = \omega - \Omega$  and  $\Lambda_1$  and  $\Lambda_2$  are the relevant contributions of spontaneous Raman scattering at these frequencies.

The expressions derived for the informative part of the spectral Mandel parameter (10) show that the most physically interesting contributions related to light scattering are partially masked by a trivial effect of linear absorption. It might seem that the contributions related to linear absorption can be easily selected since they are, in fact, independent of the beat frequency  $\Omega$ . However, this cannot be done because these contributions substantially lower the sensitivity of the sub-Poissonian modification of the IFS method [see the estimates given by Eqs. (6) and (9)]. In fact, it is easy to understand that the measurement of the spectrum of weak Raman scattering in the sub-Poissonian regime is possible only if the last two terms on the right-hand side of Eq. (10) are much larger than the first term.

To estimate these contributions, we should take into consideration the spatial properties of scattered radiation and the multimode character of the problem. Since Stokes and anti-Stokes components provide equal contributions in the equilibrium regime,  $\Lambda_1 \sim \Lambda_2 = \Lambda$ , we arrive at the following estimate:

$$\Lambda(\Omega) \approx J_p N_0 \frac{d\sigma_R}{d\mathcal{C}} \Delta\mathcal{C}_{\text{Coh}} \frac{\tau_1}{(1 + ((\Omega - \Omega_0)\tau_1)^2)}. \quad (11)$$

This estimate gives the number of photons scattered into an elementary solid angle  $\Delta\mathcal{C}_{\text{Coh}}$  per unit time per unit spectral range. Here,  $J_p$  stands for the photon fluence in the scattered wave,  $\Delta\mathcal{C}_{\text{Coh}}$  is the collection angle

of scattered radiation, and the differential cross section of Raman scattering  $d\sigma_R/d\mathcal{C}$  corresponds to an arbitrary observation direction. Thus, for the number of scatterers equal to  $N_0$  and characteristic relaxation time  $\tau_1$  determined for some resonant transition  $\Omega_0$  in the lower state set of sublevels, the right-hand side of Eq. (11) can be estimated as

$$\Lambda(\Omega) \sim J_p \lambda^2 \tau_1 N_0 \frac{1}{4\pi S_0} \sigma_R. \quad (12)$$

Here, we assumed for simplicity that scattering is isotropic and the collection angle of scattered radiation can be estimated as

$$\Delta\mathcal{C}_{\text{Coh}} \approx O(1) \frac{8\pi^2}{S_0 k^2}, \quad (13)$$

where  $S_0$  is the cross-section area of the heterodyne beam and  $\lambda = k^{-1}$  determines the heterodyne wavelength divided by  $2\pi$ .

On the other hand, the first term on the right-hand side of Eq. (10) can be estimated as

$$\kappa_1 \sim \kappa_2 \sim N_0 \frac{\sigma_0}{S_0}, \quad (14)$$

where  $\sigma_0$  is the absorption cross section. When scattering corresponding to the wings of optical transition lines are studied, the cross section of Raman scattering  $\sigma_R$  and the absorption cross section  $\sigma_0$  are approximately of the same order of magnitude. Thus, we see that the increase in the Mandel parameter due to Raman scattering plays a dominant role if  $J_p \lambda^2 \tau_1 \gg 1$ , i.e., if the photon fluence in the scattered wave is sufficiently high. The intensity of scattered wave can be, in fact, increased up to the levels where saturation effects become noticeable,  $J_p \lambda^2 \gamma^{-1} \sim 1$ , where  $\gamma^{-1}$  is the natural lifetime of the upper level. Thus, provided that  $\gamma \tau_1 \gg 1$ , the contribution of Raman scattering to the increase in the Mandel parameter, indeed, becomes predominant. This inequality is usually satisfied if the lower sublevels belong to the ground state or these sublevels are metastable.

We should emphasize two important circumstances. First, even in the case when  $J_p \lambda^2 \tau_1 \sim \gamma \tau_1 \gg 1$ , the change in the Mandel parameter with  $N_0 \sim 1$  may be small in practice due to the smallness of the ratio  $\sigma_R/S_0$ . In other words, if the number of scattering species is small, the mean number of photons scattered at a shifted frequency (per unit time per unit spectral range) into a small collection solid angle is small as compared with unity:  $\Lambda(\Omega) \ll 1$ . Second, only small values of Mandel parameter may be of interest for the above-described modification of optical mixing spectroscopy based on the scattering of radiation into modes filled with squeezed light. Indeed, if the mean number of scattered photons is higher than or on the order of unity,  $\Lambda(\Omega) \geq 1$ ,

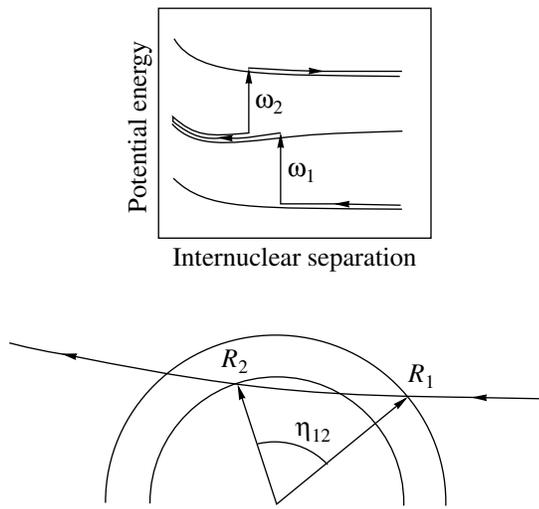
then this approach provides no improvement in sensitivity relative to the heterodyne detection of light scattered into vacuum modes. Thus, the method proposed in this paper is essentially oriented at systems with a low number of scatterers. Such systems are of considerable practical interest, for example, for biophysical applications, where a single scatterer may have a unique nature.

### 3. TWO-PHOTON POLARIZATION-DEPENDENT TRANSITIONS WITH THE USE OF NONCLASSICAL LIGHT

Another example demonstrating possible spectroscopic applications of nonclassical light is related to polarization-dependent two-photon absorption. At first glance, we should expect that it would be of interest to observe such a process for free atoms by irradiating atoms with pairs of polarization-entangled photons in a scheme that would be similar to the experimental scheme of [7] and that would generalize this scheme. However, atoms possess a high internal symmetry, and any effects related to nonclassical correlation in the polarization dependence of a two-photon process could be observed only for preliminarily polarized atoms or for atoms placed in an external field. In both cases, an atom as an absorber is similar in many of its properties to a two-photon polarization-sensitive correlation interferometer. Nonclassical polarization-dependent interferometric measurements are widely used, for example, in applications related to the problems of quantum information [16]. Therefore, a conceptually similar experiment with an atomic absorber would not be very interesting.

The situation radically changes if we deal with a two-photon excitation of a molecular or quasi-molecular system where an atom involved in optical transitions interacts with another atom, e.g., in the collision process. The symmetry of optical transitions is defined in the internal molecular system of coordinates related to the interatomic axis. In the case of experiments performed with a gas cell or an atomic trap, the orientation of the internal system of coordinates is random for each pair of colliding atoms and is unknown in experiments. If a quasi-molecule is excited by light from two independent classical sources, then the only way to study the polarization dependence of the process is to change the relative polarization of these sources defined with respect to the external laboratory system of coordinates. If the excitation photons form a polarization-entangled pair, then the polarization dependence should be observed, although no information on the polarization direction of each of the photons can be obtained (in contrast to interferometric measurements).

A typical scheme of a two-photon optical control of an atomic collision is shown in Fig. 3. We assume that pairs of photons with frequencies  $\omega_1$  and  $\omega_2$  are created in the process of spontaneous parametric scattering of the second type and are, consequently, characterized by



**Fig. 3.** Diagram illustrating the principle of optical control of an atomic collision. Optical Franck–Condon transitions are initiated at the points  $R_1$  and  $R_2$  separated along the collisional trajectory by the deviation angle of the internuclear axis  $\eta_{12}$ .

a nonclassical type of correlation of their polarizations (see [17]). The joint wave function of such photons can be represented as

$$|\Psi\rangle_{12} = \frac{1}{\sqrt{2}} [|\mathbf{e}_x\rangle_1 |\mathbf{e}_y\rangle_2 + e^{i\varphi} |\mathbf{e}_y\rangle_1 |\mathbf{e}_x\rangle_2]. \quad (15)$$

The concept of polarization defined for each of the photons does not exist in principle for a state described by such a wave function. However, the relative polarizations of these photons are strictly correlated. In the particular case of  $\varphi = \pi$ , if some of the polarization states is implemented in experiments for one of the photons, the polarization state of the second photon also becomes definite and orthogonal to the polarization of the first photon.

Let us consider, as an example, an optical collision often discussed in the literature that implements transitions on the wings of the  ${}^1S \rightarrow {}^1P \rightarrow {}^1S$  two-photon transition of one of the atoms. The second atom is assumed to remain in the ground state, in fact, playing the role of an external potential, perturbing energy levels of the first atom. Transitions of this type correspond, for example, to the case of collisions of a second-group atom with an inert atom. For simplicity, we ignore for a while the rotation of the quasi-molecule and assume that head-on collisions provide a dominant contribution to the transition probability. As shown in [13], in the Franck–Condon approximation, the cross section of optical collision determining the probability of excitation of an active atom to an upper state is characterized by the following dependence on the phase  $\varphi$  for both

possible excitation channels,  ${}^1\Sigma \rightarrow {}^1\Sigma \rightarrow {}^1\Sigma$  and  ${}^1\Sigma \rightarrow {}^1\Pi \rightarrow {}^1\Sigma$ :

$$\sigma_0 \propto w^{(1)} w^{(2)} (1 + \cos\varphi). \quad (16)$$

Here,  $w_1$  and  $w_2$  are atomic excitation probabilities at Franck–Condon transition points.

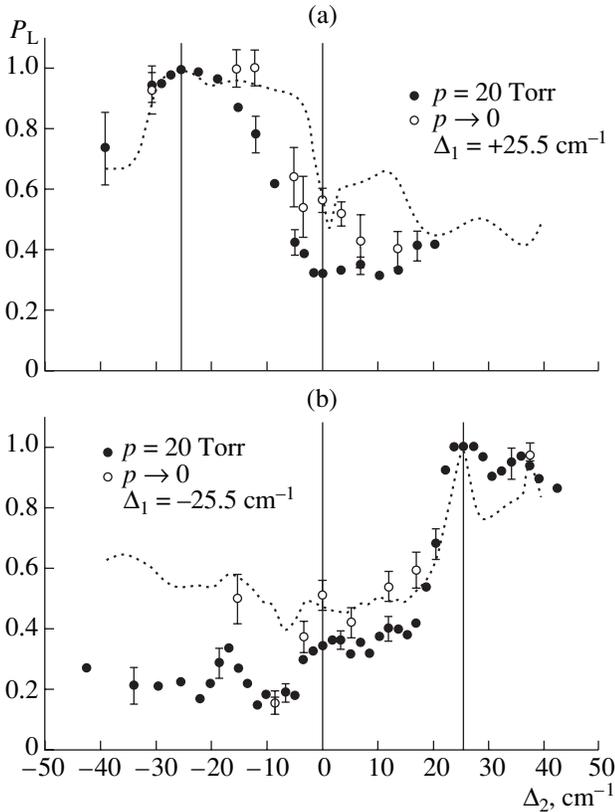
The dependence of  $\sigma_0$  on the phase  $\varphi$  indicates that quantum correlations between polarization modes of an optical parametric oscillator may influence the internal dynamics of an optically controlled collision process. If the phase  $\varphi$  approaches  $\pi$ , the transition probability considerably lowers. Such a dependence becomes clear if we consider the wave function (15), describing a polarization-entangled state of a system of two photons. As mentioned above, if  $\varphi = \pi$ , the first photon is characterized by some unknown direction of polarization, while the polarization of the second photon is always orthogonal to the polarization of the first photon. Absorption of the first photon in this case leads to a reduction of the quantum state of the entire system, fixing polarization directions of both the first and the second photons in the internal system of coordinates. Since these directions are mutually orthogonal, the second photon cannot be absorbed through two-photon transitions of the above-specified type, and such excitation channels are closed. It is clear that such an effect is impossible from the viewpoint of classical electrodynamics, since it would be impossible to prepare a light polarized along an arbitrary and unknown direction in space (or orthogonal to this arbitrary direction).

Let us consider now the results of consistent numerical simulations of the polarization dependence for a two-photon transition based on the general theory from [18] including effects related to the rotation of a quasi-molecule. These simulations will allow us to refine and test the above-presented qualitative estimate, as well as to compare polarization dependences corresponding to excitation with classical and quantum light. Let us explain, first of all, why such a comparison is possible in principle.

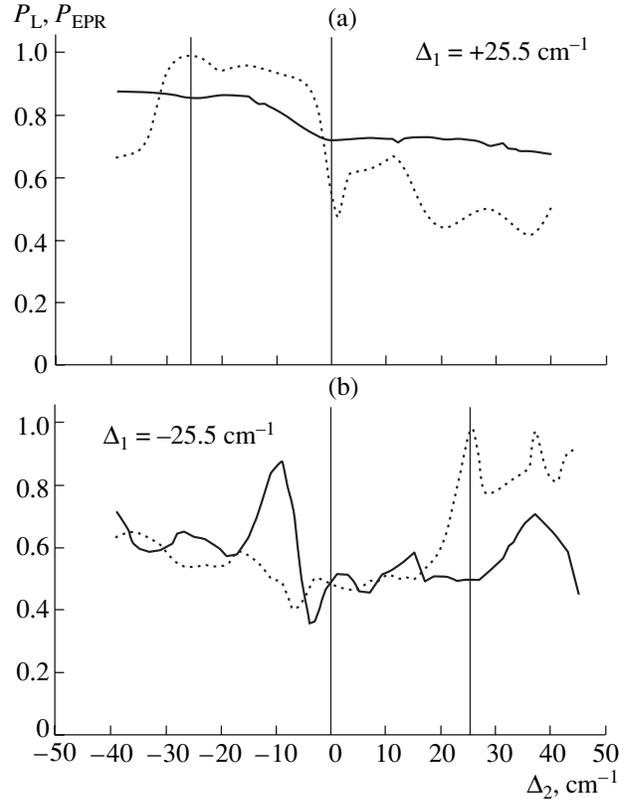
The cooperative wave function  $|\Psi\rangle_{12}$  defined by Eq. (15) can be represented in the form of another expansion in the system of coordinates where the  $\xi$ - and  $\eta$ -axes are oriented at the angle of  $\pi/4$  with respect to the  $x$ - and  $y$ -axes. It is easy to verify that, with  $\varphi = 0$ , we have

$$|\Psi\rangle_{12} = \frac{1}{\sqrt{2}} [|\mathbf{e}_\xi\rangle_1 |\mathbf{e}_\xi\rangle_2 - |\mathbf{e}_\eta\rangle_1 |\mathbf{e}_\eta\rangle_2]. \quad (17)$$

This can be considered under certain conditions (if we neglect, in fact, the interference effect) as a mutual parallel polarization of the first and second quanta. It would be reasonable to place the excitation of colliding atoms by such a pair of photons in correspondence with the excitation of classical light beams with parallel polarizations. In the opposite case of  $\varphi = \pi$ , the wave



**Fig. 4.** Polarization spectra of a two-photon Mg–Ne optical collision ( $^1S \rightarrow ^1P \rightarrow ^1S$ ). Experimental dots show the interpolation to zero neon pressure  $p$ . The dotted curve represents the results of calculations.



**Fig. 5.** Calculated polarization spectra of a Mg–Ne optical collision ( $^1S \rightarrow ^1P \rightarrow ^1S$ ): (dotted curve) “classical” polarization dependence for  $P_L$  and (solid curve) “nonclassical” polarization dependence for  $P_{EPR}$ .

function  $|\Psi\rangle_{12}$  remains invariant with respect to rotational transformations,

$$|\Psi\rangle_{12} = \frac{1}{\sqrt{2}}[|e_\xi\rangle_1|e_\eta\rangle_2 - |e_\eta\rangle_1|e_\xi\rangle_2]. \quad (18)$$

Thus, excitation by such a pair of photons can be placed in correspondence with excitation with orthogonal-polarized classical light beams.

Representing the cooperative wave function  $|\Psi\rangle_{12}$  in the form of Eqs. (17) and (18), we can define the “nonclassical” polarization ratio  $P_{EPR}$  of the following form:

$$P_{EPR} = \frac{\sigma_0|_{\varphi=0} - \sigma_0|_{\varphi=\pi}}{\sigma_0|_{\varphi=0} + \sigma_0|_{\varphi=\pi}} = P_L + \Delta P_{int}. \quad (19)$$

The latter equality shows that a “classical” contribution,  $P_L$ , and the contribution due to two-photon interference,  $\Delta P_{int}$ , can be separated in the considered polarization ratio. The classical contribution is defined by the first equality in Eq. (19), where the numerator and the denominator involve cross sections corresponding to excitation with mutually parallel-polarized or orthogonal-polarized light beams. It is of interest to numerically compare the spectral dependences for the

polarization ratios  $P_{EPR}$  and  $P_L$  for some realistic collisional system.

As an example, we have chosen a Mg–Ne pair. Experimental data on two-photon excitation with classical light through the  $3s^2\ ^1S_0 \rightarrow 3p\ ^1P_1 \rightarrow 5s\ ^1S_0$  transition are available for this pair. Figure 4 presents the experimental data and the results of our calculations for optical collisions of magnesium and neon plotted as functions of the relevant frequency detunings  $\Delta_1$  and  $\Delta_2$  at the first and second steps of photoexcitation along with the corresponding collision energies for  $E = 300$  K. The results of calculations (shown by the dotted curve) demonstrate a quite satisfactory agreement with the experimental data. Note that the method of calculations, which was described in detail in our paper [18], does not involve any fitting parameters, and the discrepancy between the results of calculations and experimental data is mainly due to the inaccurate knowledge of interaction potentials. Figure 5 reproduces the results of calculations for the “classical” polarization ratio  $P_L$  and displays the spectral dependence for the “nonclassical” ratio  $P_{EPR}$ .

Comparison of these curves shows that the considered polarization ratios generally differ from each other

quite substantially. However, within certain spectral ranges, the influence of two-photon interference is suppressed and the “classical” dependence is close to the “nonclassical” one. Another important conclusion that can be made from the analysis of these dependences is that, within certain spectral ranges, the “nonclassical” polarization approaches its limiting value,  $P_{\text{EPR}} \rightarrow 1$ . The limiting value is achieved in those cases when an optically controlled collision occurs under conditions close to the conditions corresponding to the recoil approximation. This implies the suppression of internal depolarization processes related to the rotation of a molecule in the process of collision.

#### 4. CONCLUSION

In this paper, we have provided a theoretical generalization of the IFS technique to the case when probe radiation possesses nonclassical statistical properties. It was shown that, regardless of the specific physical nature of scatterers, detection of light scattered into modes filled with squeezed radiation may offer considerable advantages. The method of optical heterodyning employed under these conditions to analyze the spectral content of scattered radiation is characterized by a higher resolution than the resolution attainable by heterodyning the light scattered into vacuum modes.

Advantages of the considered technique over the detection of light scattered into vacuum modes become especially substantial when one deals with a small number of scatterers or even a single scatterer. This provides a unique opportunity of spectroscopic selection of a small number of microscopic objects (atoms, molecules, etc.) whose Raman or Rayleigh spectra are characterized by subtle differences (such as isotopic shifts, as well as additional shifts and broadening of lines). It would be difficult to resolve such differences with the use of other high- and ultrahigh-resolution spectroscopic techniques. Note also that the observation diagram shown in Fig. 2 is, in our opinion, simple in practical implementation and can be realized and experimentally tested with currently available sources of squeezed radiation. Thus, this results may provide some theoretical background for a new direction in the development of the IFS technique, mainly as an experimental method essentially using quantum properties of interrogating radiation.

Another example considered in this paper is related to the possibility of two-photon coherent optical control of the internal dynamics of a collisional process with the use of nonclassical polarization-entangled light. Such a control permits one to close certain photoexcitation channels, leaving the other channels open even after a full averaging over the directions of motion of colliding atoms. Photoexcitation channels are defined under these conditions with respect to the internal molecular system of coordinates, which remains completely unknown in the process of experiments. Thus, we predict a rather nontrivial possibility to control elementary photochemical processes involving

small numbers of particles using the principles of quantum statistics. We have also performed a comparative analysis of classical and nonclassical polarization dependences of the process and determined the “nonclassical” polarization ratio. The main result of this analysis is that the highest control efficiency can be achieved for the case of head-on collisions, when effects of quantum correlation are less sensitive to unwanted rotational depolarization.

#### ACKNOWLEDGMENTS

This work has been reported at the seminar on quantum optics at M.V. Lomonosov Moscow State University dedicated to the memory of D.N. Klyshko in April, 2001. We are grateful to A.V. Slavgorodskii for helping us to prepare numerical simulations.

This study was partially supported by the Russian Foundation for Basic Research (project no. 01-02-17059) and by INTAS under Grant INFO 00-479. We are also grateful to the Delzell Foundation, Inc. private charity foundation for the financial support of this work.

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