Squeezing of thermal fluctuations in four-wave mixing in a Λ scheme

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We theoretically investigated the mechanism of two-mode quadrature squeezing in a regime of four-wave mixing in a Λ scheme of three-level atoms embedded in a thermal reservoir. We demonstrated that the process of nonlinear transfer of noise from the low frequency of ground state splitting to the optical frequency is significant if the number of thermal photons at the low frequency is high. We have shown that correct calculation of the two-mode squeezing level taking into account both thermal noise and distortion of dissipative properties of the thermally excited medium resulted in a simple expression for the maximum squeezing level, which is defined by the ground-state coherence decay rate and the drive-field intensity. We found the optimal conditions for squeezing, in particular, the optimal density-length product of the active medium depending on the atomic relaxation parameters and the drive-field intensity.

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I. INTRODUCTION

Since the pioneering work of Slusher *et al.* [1] many groups have demonstrated squeezing based on four-wave mixing (4WM) in atomic vapors under a variety of conditions [2–5]. A number of theoretical and experimental works [6–15] have shown that resonant 4WM based on ground-state coherence is attractive, since coherent population trapping (CPT) and electromagnetically induced transparency (EIT) can provide a small ratio of absorption coefficient with respect to a high increment of 4WM instability and, as a consequence, strong intensity correlations of generated twin beams and a high level of two-mode quadrature squeezing between opposite sidebands of the twin beams. One of the advantages of this mechanism is that 4WM naturally generates the squeezed light with a narrow frequency band that is resonant to an atomic transition, which can be applied as a quantum-information carrier interacting with a material system. Besides, in resonant schemes of 4WM a high level of squeezing can be achieved for relatively low drive powers. Noise correlations and squeezed-light generation on the basis of 4WM in sodium vapor has been experimentally demonstrated by Grove *et al.* [6]. The demonstration of −8 dB of relative intensity squeezing between probe and conjugate beams produced in a nearly copropagating nondegenerate 4WM scheme in rubidium vapor has been reported in Refs. [12,13,16].

A common problem in experiments using resonant atomic ensembles for squeezing is the processes of spontaneous emission in atoms that occur concurrently with four-wave mixing and lead to incoherent emission into the signal modes. These processes contribute to the “thermalization” of the optical state in the signal modes and degrade the squeezing [17]. Such processes take place if there are thermal excitations in the medium (if the temperature is not zero).

The generally excepted idea [12] is that the negative influence of spontaneous emission from the upper level on squeezing in 4WM based on ground-state coherence is reduced due to the EIT effect. It is intuitively based on the standard fluctuation-dissipation theorem (FDT) [18]. But the generalized fluctuation-dissipation relation obtained in Ref. [19] for a resonantly driven quantum medium makes us believe that the situation is complicated by the fact that the noise level at the optical frequency may be determined by the averaged number of thermal photons at the low frequency of ground-state splitting if the population damping rate at this frequency is not zero. This effect is caused by the parametric transfer of noise from low to high frequency in the presence of the resonant drive wave. Actually the generalized FDT [19] takes into account the process of spontaneous Raman scattering that leads to incoherent emission into the resonant signal mode.

The presence of some extra noise beyond that which is necessary to preserve the canonical commutation relation of the field has been observed in EIT delayed light experiments [20] and in experiments on the propagation of squeezed vacuum under EIT [21] and has also been associated with the processes of population exchange between ground states in theoretical treatment [22]. The calculation of spontaneous Raman noise presented in Ref. [23] demonstrates storage and retrieval of single photons in an off-resonant Raman memory scheme.

Noise scattering as a factor that can spoil squeezing in 4WM mechanisms has been pointed in Ref. [17]. However, until now the accurate theory of 4WM squeezed-light generation in thermally excited atomic vapors based not on the phenomenological approach (as in Ref. [13]), but with gain, propagation losses, and noise intrinsically included in the microscopic approach, has not been presented. Most theoretical studies [8,15] have used a zero-temperature approximation for the analysis of squeezed-light generation in such schemes. In most of works the theoretical treatment has been limited to numerical calculations without analytical solutions, clarification of basic dependencies, and optimal conditions.

In the present paper we theoretically investigate the process of generation of a two-mode squeezed vacuum based on a 4WM mechanism realized in a three-level Λ scheme with monochromatic driving under the condition of thermal excitations. The considered system is interesting also from the methodological point of view as a spectacular example that illustrates the mechanisms of transformation of the field noise due to the self-consistent parametric interaction in resonant nonlinear medium.

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It should be pointed that the 4WM squeezing experiments and the experiments on paired photon generation in both "cold" [9,10] and "hot" [13] atomic vapors were performed by different groups. The main advantage of the cold conditions is that the decoherence processes are essentially reduced, while the hot conditions are attractive due to large optical depths. In the present paper we analyze mainly how squeezing depends on thermal population distribution and corresponding spontaneous emission processes. Meanwhile we obtain a useful expression for the level of squeezing depending on relaxation rates and the density of the atomic sample; the specific temperature dependencies of these parameters are very important but their analysis is beyond the scope of the present investigation.

In the present paper we use the results of previous research on this system [24] conducted for the case of zero temperature and negligible dissipation, as well as the results of analysis of this scheme of parametric instability for classical fields [25,26].

In Sec. II we introduce with the necessary degree of detail the basic terms and equations used for the analysis of quantum radiation propagating through the dense medium consisting of independent quantum centers ("atoms") interacting with a dissipative reservoir under the condition of nonzero temperature. In Sec. III the system of resonant 4WM in a Λ scheme is described. The characteristics of the parametric interaction of waves taking into account the thermal redistribution of populations among energy levels are obtained. On the basis of the approach developed in Ref. [19] the correlation relations for the noise sources of interacting quantum fields are derived. In Sec. IV the regime of two-mode squeezed-vacuum generation in this system is analyzed. The spectral properties of the squeezed vacuum under different conditions are investigated. The optimal parameters for squeezing are obtained.

II. GENERAL EQUATIONS

Here we introduce in detail the basic terms and relations for the system of quantum radiation interacting with the medium consisting of quantum atoms embedded in a dissipative reservoir in the frame of the Heisenberg-Langevin approach.

A. Atomic system

For the atomic system we define the coordinate-dependent density operator as

$$\hat{\rho}_{mn}(\mathbf{r},t) = \frac{1}{\Delta V_r}\sum_j \hat{\rho}_{j;mn}(t), \tag{1}$$

where index j numerates the atoms within the small volume ΔV_r in the vicinity of the point with the radius-vector $\mathbf{r}$ and $\hat{\rho}_{j;mn} = \hat{a}^{\dagger}_{j;n}\hat{a}_{j;m}$ is the Heisenberg density operator acting on variables of the atom with index j; it is expressed via the creation and annihilation operators which are defined by the expressions $\hat{a}^{\dagger}_n|0\rangle = |n\rangle$ and $\hat{a}_n|n\rangle = |0\rangle$, where $|n\rangle$ are basis states of the single-particle Hamiltonian with energy levels W_n. The operator $\hat{\rho}_{mn}$ obeys the Heisenberg-Langevin equation [27,28]:

$$\dot{\hat{\rho}}_{mn} = -\frac{i}{\hbar}(\hat{h}_{mp}\hat{\rho}_{pn} - \hat{\rho}_{mp}\hat{h}_{pn}) + \hat{R}_{mn} + \hat{F}_{mn}, \tag{2}$$

where $\hat{h}_{mn} = W_m\delta_{mn} - \mathbf{d}_{mn}\hat{\mathbf{E}}(\mathbf{r},t)$ takes into account the interaction of atoms with the electric field $\hat{\mathbf{E}}(\mathbf{r},t)$ in the dipole approximation, $\mathbf{d}_{mn}$ is the dipole matrix elements, $\hat{R}_{mn}$ is the relaxation operator, and $\hat{F}_{mn}$ is the Langevin noise operator satisfying $\hat{F}_{mn} = \hat{F}^{\dagger}_{nm}$, $\langle\hat{F}_{mn}\rangle = 0$; hereinafter averaging is taken over the reservoir variables and the atomic state.

The standard model for the relaxation operators corresponds to the so-called master equations [29–31], where

$$\hat{R}_{mn} = \sum_{pq} r_{mnpq}\hat{\rho}_{pq}. \tag{3}$$

In a simplest form the nonzero coefficients r_{mnpq} set the rates of transverse and longitudinal relaxation:

$$\hat{R}_{mn} = -\gamma_{mn}\hat{\rho}_{mn},\ m \neq n,$$
$$\hat{R}_{mm} = \sum_n w_{mn}\hat{\rho}_{nn}. \tag{4}$$

The correlation functions for the atomic noise operators are derived in the Appendix. Here we use the correlation relations for the spectral components of the Langevin operators, defined as $\hat{F}_{mn}(\mathbf{r},t) = \int_{-\infty}^{+\infty}\hat{F}_{mn}(\mathbf{r},\omega)e^{-i\omega t}d\omega$. Under the adiabatic approximation neglecting the slow evolution of populations $\langle\hat{\rho}_{nn}\rangle$ and the amplitude of drive-induced coherence at the resonant transition σ_{ba}, defined as $\langle\hat{\rho}_{ba}\rangle = \sigma_{ba}e^{\mp i\omega_d t}|_{b\gtrless a}$, we get

$$\langle\hat{F}_{mn}(\mathbf{r},\omega)\hat{F}_{nm}(\mathbf{r}',\omega')\rangle = \frac{1}{2\pi}(2\gamma_{mn}\langle\hat{\rho}_{nn}\rangle + \langle\hat{R}_{nn}\rangle)\delta(\omega+\omega')\delta(\mathbf{r}-\mathbf{r}'), \tag{5}$$

$$\langle\hat{F}_{ma}(\mathbf{r},\omega)\hat{F}_{bm}(\mathbf{r}',\omega')\rangle = \frac{1}{2\pi}(\gamma_{am}+\gamma_{bm}-\gamma_{ab})\sigma_{ba}\delta(\omega+\omega'\mp\omega_d)|_{b\gtrless a}\delta(\mathbf{r}-\mathbf{r}'). \tag{6}$$

B. The field equations

The electric field operator $\hat{\mathbf{E}}(\mathbf{r},t)$ in a general case obeys the following operator wave equation [27,30]:

$$\frac{\partial^2}{\partial t^2}(\hat{\mathbf{E}} + 4\pi\hat{\mathbf{P}}) + c^2\nabla\times\nabla\times\hat{\mathbf{E}} = 0, \tag{7}$$

where the operator of electric polarization $\hat{\mathbf{P}}(\mathbf{r},t) = \sum_{m,n}\mathbf{d}_{nm}\hat{\rho}_{mn}$ is expressed via density operators $\hat{\rho}_{mn}$, which are the solution of Eq. (2). Assume the field to be quantized over spatial modes in homogeneous, dissipationless medium with linear dielectric permittivity $\epsilon(\omega) = 1 + \int_0^{\infty}4\pi\chi^H(\tau)e^{i\omega\tau}d\tau$, whereas the nonlinear response, dissipation, and noise effects are taken into account as small additional terms to the linear relation:

$$\hat{\mathbf{P}}(\mathbf{r},t) = \int_0^{\infty}\chi^H(\tau)\hat{\mathbf{E}}(\mathbf{r},t-\tau)d\tau + \delta\hat{\mathbf{P}}. \tag{8}$$

Here χ^H is the Hermitian part of the linear susceptibility of the medium. We assume that the field consists of one or several quasimonochromatic waves labeled by index j; each of them may be presented as a combination of large numbers of spectrally close modes of quantization propagating within a paraxial beam of the cross-sectional area $S_\perp$. Then the following representation can be used:

$$\hat{\mathbf{E}}(\mathbf{r},t) = \sum_j \mathbf{e_j} E_j \hat{c}_j(\mathbf{r},t) e^{i\mathbf{k_j r}-i\omega_j t} + \mathbf{e_j}^* E_j \hat{c}_j^\dagger(\mathbf{r},t) e^{-i\mathbf{k_j r}+i\omega_j t},$$

where $\mathbf{e_j}$ is the unit vector of the polarization of the j wave, ω_j and $\mathbf{k_j}$ are coupled by the corresponding dispersion relation $k_j^2 c^2/\omega_j^2 = \epsilon(\omega_j)$, and operators $\hat{c}_j(\mathbf{r},t)$ and $\hat{c}_j^\dagger(\mathbf{r},t)$ are slow time- and space-dependent photon annihilation and creation operators. E_j are normalization constants defined as $E_j = \sqrt{4\pi\hbar\omega_j^2/|\frac{\partial(\omega_j^2\epsilon(\omega_j))}{\partial\omega_j}|}$ [27,30]. For such representation the operators $\hat{n}_{\mathrm{ph}j} = \hat{c}_j^\dagger(\mathbf{r},t)\hat{c}_j(\mathbf{r},t)$ play the role of the photon density operators and $\hat{\mathbf{p}}_{\mathrm{ph}j} = \mathbf{v}_{\mathrm{gr}j}\hat{c}_j^\dagger(\mathbf{r},t)\hat{c}_j(\mathbf{r},t)$ are the photon flux density operators, where $\mathbf{v}_{\mathrm{gr}j} = 2c^2\mathbf{k_j}/\frac{\partial(\omega_j^2\epsilon)}{\partial\omega_j}$ are the group velocities.

The following truncated equations can be written for the operators $\hat{c}_j(\mathbf{r},t)$ [24,28]:

$$\left(\frac{\partial}{\partial t} + (\mathbf{v}_{\mathrm{gr}j}\nabla)\right)\hat{c}_j(\mathbf{r},t) = \Lambda_j \frac{i}{\hbar}\mathbf{e}^* E_j \delta\hat{\mathbf{P}}_j(\mathbf{r},t), \qquad (9)$$

where $\Lambda_j = \mathrm{sgn}(\frac{\partial(\omega_j^2\epsilon)}{\partial\omega_j})$ and $\delta\hat{\mathbf{P}}_j(\mathbf{r},t)$ are the slowly varying amplitudes of the polarization terms $\delta\hat{\mathbf{P}} = \sum_j \delta\hat{\mathbf{P}}_j(\mathbf{r},t)e^{i\mathbf{k_j r}-i\omega_j t} + \delta\hat{\mathbf{P}}_j^\dagger(\mathbf{r},t)e^{-i\mathbf{k_j r}+i\omega_j t}$.

In a boundary-value problem the spectral decomposition of the field amplitude is used:

$$\hat{c}_j(\mathbf{r},t) = \int_{-\infty}^{+\infty} \hat{c}_{j_\nu}(\mathbf{r})e^{-i\nu t}d\nu, \qquad (10)$$

where the integration in infinite limits is reduced to the integration over the narrow frequency interval $\Delta\nu_j \ll \omega_j$. It is appropriate to write the propagation equation for the "flux" amplitudes $\hat{a}_j = \sqrt{|v_{\mathrm{gr}j}|}\hat{c}_j$. Assuming that the wave propagates in the positive z direction, $\mathbf{k_j} = k_j\mathbf{z_0}$, we get the following equation:

$$\frac{\partial\hat{a}_{j_\nu}(z)}{\partial z} - \frac{i\nu}{v_{\mathrm{gr}j}}\hat{a}_{j_\nu}(z) = \frac{i\mathbf{e}^* E_j}{\hbar\sqrt{|v_{\mathrm{gr}j}|}}\delta\hat{\mathbf{P}}_{j_\nu}(z), \qquad (11)$$

where $v_{\mathrm{gr}j} = 2c^2k_j/\frac{\partial(\omega_j^2\epsilon)}{\partial\omega_j}$ and $\delta\hat{\mathbf{P}}_{j_\nu}(\mathbf{r})$ is the corresponding spectral image of the slowly varying amplitude of polarization:

$$\delta\hat{\mathbf{P}}_j(\mathbf{r},t) = \int_{-\infty}^{+\infty} \delta\hat{\mathbf{P}}_{j_\nu}(\mathbf{r})e^{-i\nu t}d\nu. \qquad (12)$$

At the boundary z_b between the medium and vacuum the following boundary condition for the field operator can be used:

$$\frac{1}{\sqrt{c}}\hat{a}_j(z_b)\mid_{\mathrm{medium}} = \hat{c}_j(z_b)\mid_{\mathrm{vacuum}}, \qquad (13)$$

which satisfies the conservation of Poynting flux. (It is assumed that the effects of reflection are neglected.)

The polarization on the right-hand side of Eq. (11) includes nonlinearity (e.g., parametric coupling of different waves), dissipation, and fluctuations: $\delta\hat{\mathbf{P}}_{j_\nu} = \delta\hat{\mathbf{P}}_{j_\nu}^N + \delta\hat{\mathbf{P}}_{j_\nu}^{\mathrm{diss}} + \delta\hat{\mathbf{P}}_{j_\nu}^L$. The dissipation term that is linear over the quantum field, $\delta\hat{\mathbf{P}}_{j_\nu}^{\mathrm{diss}} = \chi^{\mathrm{aH}}(\omega_j+\nu)\mathbf{e}E_j\hat{c}_{j_\nu}$, defines the absorption coefficient $\kappa_j(\nu) = -i\frac{2\pi k_j}{\epsilon_j}\chi^{\mathrm{aH}}(\omega_j+\nu)$. Here χ^{aH} is the anti-Hermitian part of the susceptibility. Note that if the medium is driven by a classical field then the "linear" coefficients take into account dependence on drive intensity: $\epsilon(I_d)$, $\chi^{\mathrm{aH}}(I_d)$, and $\kappa_j(I_d)$.

So we use the following form of equation for the filed operator:

$$\frac{\partial\hat{a}_{j_\nu}(z)}{\partial z} - \frac{i\nu}{v_{\mathrm{gr}j}}\hat{a}_{j_\nu}(z) + \kappa_j(\nu)\hat{a}_{j_\nu}(z) = \hat{N}_{j_\nu} + \hat{L}_{j_\nu}. \qquad (14)$$

Here $\hat{N}_{j_\nu} = \frac{i\mathbf{e}^* E_j}{\hbar\sqrt{|v_{\mathrm{gr}j}|}}\delta\hat{\mathbf{P}}_{j_\nu}^N$ and $\hat{L}_{j_\nu} = \frac{i\mathbf{e}^* E_j}{\hbar\sqrt{|v_{\mathrm{gr}j}|}}\delta\hat{\mathbf{P}}_{j_\nu}^L$.

The Langevin term $\hat{L}_{j_\nu}$ provides the fulfillment of commutation relations for the field operators in a dissipative medium [24,28]:

$$\left[\hat{a}_{j_\nu}(z),\hat{a}_{j_{\nu'}}^\dagger(z)\right] = \frac{1}{2\pi S_\perp}\delta(\nu-\nu').$$

So the definite relation between noise terms and the absorption coefficient follows from this general requirement without specification of the dissipation and noise origin:

$$\left[\hat{L}_{j_\nu}(z),\hat{L}_{j_{\nu'}}^\dagger(z')\right] = \frac{1}{\pi S_\perp}\kappa_j(\nu)\delta(z-z')\delta(\nu-\nu'), \qquad (15)$$

but the separate expressions for the correlation functions of Langevin operators $\langle\hat{L}_{j_\nu}(z)\hat{L}_{j_{\nu'}}^\dagger(z')\rangle$ and $\langle\hat{L}_{j_{\nu'}}^\dagger(z')\hat{L}_{j_\nu}(z)\rangle$ cannot be evaluated from this relation. Their correct calculation should be based on the "microscopic" approach, by consistent calculation of noise atomic response on the action of atomic noise operators $\hat{F}_{mn}$, obeying correlation relations (5) and (6), and corresponding evaluation of the noise component of medium polarization.

III. RESONANT FOUR-WAVE MIXING IN A Λ SCHEME

A. The key parameters of the model

We consider the following scheme of the four-wave mixing process, depicted in Fig. 1, where the total field consists of the strong classical drive wave and signal quantum waves, the probe wave and the Stokes wave, copropagating in the z direction through the plane-parallel layer of three-level atoms:

$$\hat{\mathbf{E}} = \mathbf{e_d}\xi_d e^{ik_d z - i\omega_d t} + \mathrm{c.c.} + \mathbf{e_p}E_p\hat{c}_p(z,t)e^{ik_p z - i\omega_p t} + \mathbf{e_s}E_s\hat{c}_s(z,t)e^{ik_s z - i\omega_s t} + \mathrm{H.c.} \qquad (16)$$

For simplicity we put $\mathbf{e_d} = \mathbf{e_p} = \mathbf{e_s} = \mathbf{e}$. We consider the classical wave with constant amplitude $\xi_d = |\xi_d|^{i\theta}$ and a frequency equal to the frequency of the atomic transition $|2\rangle - |3\rangle$: $\omega_d = \omega_{32}$. The spectral decomposition of the wave amplitudes is used [Eq. (10)]:

$$\hat{c}_{p,s}(\mathbf{r},t) = \int_{-\infty}^{+\infty} \hat{c}_{p,s_\nu}(\mathbf{r})e^{-i\nu t}d\nu. \qquad (17)$$

Summarizing the results of previous investigations of this system [24,25] we can get that the effective generation of the bichromatic correlated radiation (and the squeezed two-mode

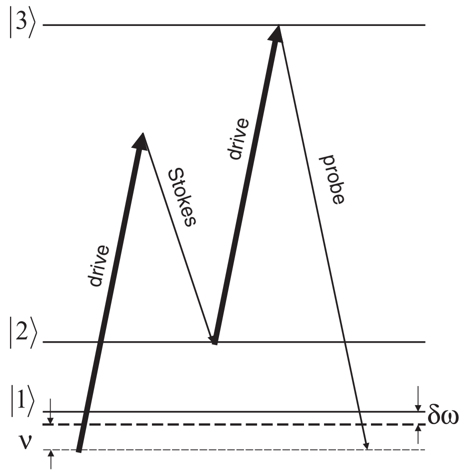

FIG. 1. The scheme of the resonant 4WM process in three-level Λ atoms. Here $\delta\omega = \omega_p - \omega_{31}$ is the one-photon resonance detuning of the "central" frequency of the quasimonochromatic probe wave, and $\delta\omega$ is chosen so that it corresponds to the strict four-wave spacial synchronism. ν is the detuning of the considered spectral component of the probe wave from the "central" frequency.

radiation) at the frequencies obeying the four-wave resonance condition,

$$\omega_p + \omega_s = 2\omega_d, \tag{18}$$

is realized due to the combination of the following factors

(i) Electromagnetically induced transparency. The drive wave should be strong enough but can be far from saturation value:

$$\gamma_{31}^2 \gg |\Omega|^2 \gg \gamma_{31}\gamma_{21}, \tag{19}$$

where $\Omega = (\mathbf{d_{32}e_d}\xi_d)/2\hbar$ is the Rabi frequency of the drive wave and γ_{ij} are the coherence decay rates at the corresponding atomic transitions. The frequency of the probe wave should be close to the two-photon resonance ($\omega_p - \omega_d \approx \omega_{21}$), which in our case corresponds to the resonance with the atomic transition $|1\rangle - |3\rangle$, adjacent to the transition $|1\rangle - |2\rangle$ with long-lived coherence $\gamma_{21} \ll \gamma_{31},\gamma_{32}$. The width of the EIT window is defined by the condition $|\omega_p - \omega_{31}| < \Delta_{\rm EIT}$, where $\Delta_{\rm EIT} \sim |\Omega|^2/\gamma_{31}$. The EIT condition, Eq. (19), provides both sufficient reduction of the partial decrement of the resonant probe wave and strong parametric coupling for configuration with close lower atomic levels:

$$\omega_{21} \ll \omega_{31},\omega_{32}. \tag{20}$$

(ii) Four-wave spacial synchronism. This condition is provided by favorable wave dispersion in the considered atomic medium. We suppose that the "central" frequencies of the probe and Stokes waves correspond to the strict four-wave spacial synchronism:

$$k_p + k_s = 2k_d, \tag{21}$$

where $k_j = (\omega_j/c)\sqrt{\epsilon(\omega_j)}$. It was shown in Refs. [25,26] that the relation (21) is fulfilled for definite detuning from the resonance with the atomic transition $\omega_p = \omega_{31} + \delta\omega$, where

$$\delta\omega = \frac{|\Omega|^2}{\omega_{21}}\left(\frac{3}{2} + |\zeta|^2\right), \tag{22}$$

here $\zeta = \frac{\mathbf{d_{31}e}}{\mathbf{d_{32}e}}$.

The effective generation of correlated radiation is realized if the parametric coupling coefficient is large compared with the partial decrement of the probe wave, which is reduced to the following relation:

$$\frac{|\Omega|^2}{\gamma_{21}\omega_{21}} > 1. \tag{23}$$

The smallness of the synchronism shift compared to the parametric coupling coefficient determines the frequency band of parametric instability near the central frequency $|\nu| < \Delta_P$, where

$$\Delta_P = \frac{2|\Omega|^2}{\omega_{21}}. \tag{24}$$

The regime of two-mode squeezed-vacuum generation in this system was analyzed in detail in Ref. [24] for the case of thermally unexcited atoms and negligible dissipation. Our aim is to analyze this regime in a dissipative atomic medium in a "hot" reservoir, so we use the results of Ref. [24] where they are applicable only.

Note that here we analyze the particular case of zero one-photon detuning for the drive wave $\omega_d = \omega_{32}$. At the same time for a number of the experiments [12–14,16] all the fields are detuned from the exact transitions to minimize the optical losses and detrimental effects of spontaneous emission. From this point of view the analyzed system can be considered as a demonstrative one that enables us to estimate the maximal negative influence of spontaneous emission to squeezing. On the other hand this system is interesting since the strict one-photon resonance provides the largest nonlinear coupling coefficient as one of two competing parameters together with optical losses. The schemes with strict resonance were realized in other experiments [6,9,10]. Developing the theory for any detunings as well as for Doppler broadening is the plan for future investigation.

B. Equations for the density-matrix operators

Here we write the equations for the density-matrix operators, Eq. (2), in the presence of the electric field, Eq. (16), and Langevin forces $\hat{F}_{mn}$ in order to calculate the coherently driven partial, parametric, and noise components of polarization. Using the rotating-wave approximation (RWA) with respect to the high frequencies ω_{31} and ω_{32} under the condition of Eq. (20) the interaction of all field components with both high-frequency transitions is taken into account, so that the density-matrix operators are presented as

$$\hat{\rho}_{31} = \sum_{j=d,p,s} \hat{\sigma}_{31}^j e^{-i\omega_j t + ik_j z}, \quad \hat{\rho}_{32} = \sum_{j=d,p,s} \hat{\sigma}_{32}^j e^{-i\omega_j t + ik_j z},$$

$$\hat{\rho}_{21} = \hat{\sigma}_{21} e^{-i\omega_l t + ik_l z},$$

where $\omega_l = \omega_p - \omega_d$ and $k_l = k_p - k_d$. Meanwhile, we use the approximation of well-resolved transitions:

$$\gamma_{31},\gamma_{32} \ll \omega_{21}. \tag{25}$$

We also assume the following condition to be fulfilled:

$$\frac{\gamma_{31}|\Omega|^2}{\gamma_{21}\omega_{21}^2} \ll 1. \tag{26}$$

As it will be seen later, the condition of Eq. (26) enables us to neglect the partial dissipation of the nonresonant Stokes wave with respect to the dissipation of the resonant probe wave. It is fulfilled if the small relaxation rate at transition $|1\rangle - |2\rangle$ is taken into account.

Consider the atomic response to the action of quantum radiation and noise forces additively in a linear approximation over quantum fields. Assume that the amplitude of the classical drive wave is constant. Working in spectral representation for the field operators, Eq. (10), we use the corresponding representation for the slow components of the density-matrix operators:

$$\hat{\sigma}^{p,s}_{31,32}(z,t) = \int_{-\infty}^{+\infty} \hat{\sigma}^{p,s}_{31,32_\nu}(z)e^{-i\nu t}d\nu,$$
$$\hat{\sigma}_{21}(z,t) = \int_{-\infty}^{+\infty} \hat{\sigma}_{21_\nu}(z)e^{-i\nu t}d\nu,$$

where the width of corresponding spectral lines is small compared to the atomic frequency ω_{21}. Similarly the spectrum of Langevin forces $F_{mn}(z,\omega)$ can be separated into three intervals:

$$\hat{F}_{31,32}(z,\omega) = \sum_{j=p,s} \hat{f}^{j}_{31,32_{\nu=\omega-\omega_j}}(z),$$
$$\hat{F}_{21}(\omega,z) = \hat{f}_{21_{\nu=\omega-\omega_l}}(z). \tag{27}$$

Then the system of equations for the density-matrix operators takes the following form of algebraic equations:

$$\hat{\sigma}^{p}_{31_\nu} = \frac{1}{\Delta^{p}_{31}}\big(\hat{p}_\nu(\rho_{11}-\rho_{33}) + \Omega\hat{\sigma}_{21_\nu} + \hat{f}^{p}_{31_\nu}\big),$$
$$\hat{\sigma}^{s}_{31_\nu} = \frac{1}{\Delta^{s}_{31}}\big(\zeta\hat{s}_\nu(\rho_{11}-\rho_{33}) + \hat{f}^{s}_{31_\nu}\big),$$
$$\hat{\sigma}^{p}_{32_\nu} = \frac{1}{\Delta^{p}_{32}}\left(\frac{1}{\zeta}\hat{p}_\nu(\rho_{22}-\rho_{33}) + \hat{f}^{p}_{32_\nu}\right),$$
$$\hat{\sigma}^{s}_{32_\nu} = \frac{1}{\Delta^{s}_{32}}\big(\hat{s}_\nu(\rho_{22}-\rho_{33}) + \zeta\Omega\big(\hat{\sigma}_{21_{-\nu}}\big)^\dagger + \hat{f}^{s}_{32_\nu}\big),$$
$$\hat{\sigma}_{21_\nu} = \frac{1}{\Delta^{l}_{21}}\big(\hat{\sigma}^{p}_{31_\nu}\Omega^* + \sigma^{d}_{31}(\hat{s}_{-\nu})^\dagger - \hat{p}_\nu\sigma^{d*}_{32} - \zeta\Omega\big(\hat{\sigma}^{s}_{32_{-\nu}}\big)^\dagger + \hat{f}_{21_\nu}\big). \tag{28}$$

Here $\zeta = \frac{\mathbf{d_{31}e}}{\mathbf{d_{32}e}}$, as was introduced in Sec. III A. The field operators are

$$\hat{p}_\nu = \frac{\mathbf{d_{31}e}E_p\hat{c}_{p_\nu}}{2\hbar}, \quad \hat{s}_\nu = \frac{\mathbf{d_{32}e}E_s\hat{c}_{s_\nu}}{2\hbar}.$$

The complex frequency detunings are

$$\Delta^{j}_{mn} = \omega_{mn} - \omega_j - \nu - i\gamma_{mn}, \quad j = d,l,p,s. \tag{29}$$

For the averaged drive-induced coherences we have the following relations:

$$\sigma^{d}_{31} = \frac{1}{\Delta^{d}_{31}}\zeta\Omega(\rho_{11}-\rho_{33}),$$
$$\sigma^{d}_{32} = \frac{1}{\Delta^{d}_{32}}\Omega(\rho_{22}-\rho_{33}), \tag{30}$$

where $\Delta^{d}_{31} \approx \omega_{21}$, $\Delta^{d}_{32} = -i\gamma_{32}$. The averaged values for diagonal density-matrix operators obey the following equations:

$$-(w_{21}+w_{31})\rho_{11} + w_{12}\rho_{22} + w_{13}\rho_{33} = 0,$$
$$-2\mathrm{Im}\big(\Omega^*\sigma^{d}_{32}\big) + w_{21}\rho_{11} - (w_{12}+w_{32})\rho_{22} + w_{23}\rho_{33} = 0. \tag{31}$$

In Eqs. (28)–(31) the relaxation model, Eq. (4), is used.

C. Field-induced population redistribution

Under the condition of finite temperature $T \neq 0$ and not ideal low-frequency transition $|2\rangle - |1\rangle$, it is necessary to calculate the redistribution of atoms over levels induced by the drive wave. To this end we express the relaxation rates w_{mn}, Eq. (31), in terms of equilibrium (in the absence of drive) population distribution,

$$r^T_n = \rho^T_{nn}/\rho^T_{11} = \exp\left(-\hbar\omega_{n1}/T\right),$$

and longitudinal relaxation times, defined for transition $|m\rangle - |n\rangle$ as T_{mn}, so that

$$w_{1n} = \frac{1}{T_{n1}},\ w_{n1} = \frac{r^T_n}{T_{n1}},\ w_{23} = \frac{1}{T_{32}},\ w_{32} = \frac{1}{T_{32}}\frac{r^T_3}{r^T_2}. \tag{32}$$

We get from Eqs. (31)–(32) the following expressions for the stationary populations:

$$\frac{\rho_{nn}}{\rho_{11}} = \frac{r^T_n\left(\frac{1}{T_{21}T_{31}} + \frac{1}{T_{21}T_{32}} + \frac{1}{T_{31}T_{32}}\frac{r^T_3}{r^T_2}\right) + \frac{2|\Omega|^2}{\gamma_{32}}\left(\frac{r^T_2}{T_{21}} + \frac{r^T_3}{T_{31}}\right)}{\left(\frac{1}{T_{21}T_{31}} + \frac{1}{T_{21}T_{32}} + \frac{1}{T_{31}T_{32}}\frac{r^T_3}{r^T_2}\right) + \frac{2|\Omega|^2}{\gamma_{32}}\left(\frac{1}{T_{21}} + \frac{1}{T_{31}}\right)},$$
$$n = 2,3. \tag{33}$$

Consider the condition of EIT, Eq. (19), that is realized if $1/T_{21} \ll 1/T_{31}$ and $1/T_{32}$ and if the level of thermal excitations at optical frequencies is low, $T \ll \hbar\omega_{31}$. We put the exact equality

$$r^T_3 = 0,$$

that corresponds to real conditions with good accuracy. Meanwhile the ground-state frequency splitting can be much less than the energy of thermal excitations (for example, in Rb, where $\omega_{21} = 2\pi \times 6.83$ GHz we get $\hbar\omega_{21} \sim T$ at temperature $T \sim 0.3$ K). It is remarkable that according to Eq. (33) under the EIT condition level $|2\rangle$ is devastated due to the action of the strong drive wave and fast relaxation from the upper level, which is a manifestation of the well-known CPT effect:

$$\frac{\rho_{22}}{\rho_{11}} \approx r^T_2\left(1 + \frac{T_{31}}{T_{32}}\right)\frac{\gamma_{32}}{2T_{21}|\Omega|^2} \ll r^T_2. \tag{34}$$

Nevertheless the residual population of level $|2\rangle$ is taken into account, and, as it will be shown later, it is necessary to do this for correct noise estimation.

Note that the upper level is slightly populated due to the drive field action (although the number of atoms at the upper level does not depend on the drive intensity under the EIT condition):

$$\frac{\rho_{33}}{\rho_{11}} \approx r_2^T \frac{T_{31}}{T_{21}} \ll \frac{\rho_{22}}{\rho_{11}}, \tag{35}$$

but this population can be ignored.

D. Susceptibility of thermally excited atoms

The amplitudes of the slowly varying components of polarization $\hat{\mathbf{P}} = \sum_{j=d,p,s} (\hat{\mathbf{P}}_j(z,t)e^{ik_jz-i\omega_jt} + \hat{\mathbf{P}}_j^\dagger(z,t)e^{-ik_jz+i\omega_jt})$ are expressed via density components in the following way:

$$\hat{\mathbf{P}}_j = \mathbf{d}_{23}\hat{\sigma}_{32}^j + \mathbf{d}_{13}\hat{\sigma}_{31}^j. \tag{36}$$

We solve the dynamical part of equations for the atomic coherences, Eqs. (28), taking into account the small population of level $|2\rangle$ [the population of level $|3\rangle$ can be ignored as dictated by Eq. (35)]. As a result, we calculate the susceptibility components $\chi_p^{H,aH}$, $\chi_s^{H,aH}$, χ_{ps}, and χ_{sp} defined by the following relations:

$$\begin{aligned}\hat{P}_{p_\nu} &= \chi_p^H(\nu)E_p\hat{c}_{p_\nu} + \chi_p^{\mathrm{aH}}(\nu)E_p\hat{c}_{p_\nu} \\ &\quad + \chi_{ps}(\nu)e^{i2\theta}E_s(\hat{c}_{s_{-\nu}})^\dagger + \delta P_{p_\nu}^L, \\ \hat{P}_{s_\nu} &= \chi_s^H(\nu)E_s\hat{c}_{s_\nu} + \chi_s^{\mathrm{aH}}(\nu)E_s\hat{c}_{s_\nu} \\ &\quad + \chi_{sp}(\nu)e^{i2\theta}E_p(\hat{c}_{p_{-\nu}})^\dagger + \delta P_{s_\nu}^L.\end{aligned} \tag{37}$$

The following expressions are obtained:

$$\begin{aligned}\chi_p^H(\nu) &= \frac{\eta}{4\pi|\Omega|}\left(\frac{\omega_p+\nu-\omega_{31}}{|\Omega|} - |\zeta|^2\frac{|\Omega|}{\omega_{21}}\right), \\ \chi_p^{\mathrm{aH}}(\nu) &= \frac{i\eta}{4\pi|\Omega|}\left(\frac{(\omega_p+\nu-\omega_{31})^2\gamma_{31}}{|\Omega|^3} + \frac{\gamma_{21}}{|\Omega|} - \frac{|\Omega|}{\gamma_{32}}\frac{n_{23}}{\rho_{11}}\right) \\ &\approx \frac{i\eta}{4\pi|\Omega|}\left(\frac{\nu^2\gamma_{31}}{|\Omega|^3} + \frac{\gamma_{21}}{|\Omega|} - \frac{|\Omega|}{\gamma_{32}}\frac{n_{23}}{\rho_{11}}\right), \\ \chi_s^H(\nu) &= \frac{\eta}{8\pi\omega_{21}}\left(1 - \frac{3}{2}\frac{\omega_s+\nu-2\omega_{32}+\omega_{31}}{\omega_{21}}\right), \\ \chi_s^{\mathrm{aH}}(\nu) &= \frac{i\eta}{8\pi\omega_{21}}\left(-\frac{3}{2}\frac{\gamma_{31}}{\omega_{21}} + 2\frac{1}{\omega_{21}}\left(\gamma_{31} + \frac{\gamma_{32}}{|\zeta|^2}\right)\frac{n_{23}}{\rho_{11}}\right), \\ \chi_{ps}(\nu) &= \chi_{sp}(\nu) = -\frac{\eta}{4\pi\omega_{21}}.\end{aligned} \tag{38}$$

Here

$$\eta = 4\pi|d_{31}|^2N/\hbar, \tag{39}$$

where N is the density of atoms. The relations (38) correctly describe the frequency dependence of the susceptibility components in a central part of the EIT window, more precisely for $|\nu| < |\Omega|\sqrt{\gamma_{21}/\gamma_{31}}$. The small corrections of the order $\gamma_{31}|\Omega|^2/(\gamma_{21}\omega_{21}^2) \ll 1$ [Eq. (26)] are not taken into account. In particular this means that the frequency dependence of the anti-Hermitian part of the probe wave susceptibility $\chi_p^{\mathrm{aH}}(\nu)$ is essential only at the scale larger than the frequency band of the parametric instability $|\nu| \gtrsim |\Omega|^2/\omega_{21}$ (see Sec. III A).

In correspondence to the obtained relation for the anti-Hermitian part of partial susceptibility of the probe wave, Eq. (38), it is decreased in the presence of the population of level $|2\rangle$. To analyze how strong this reduction is, we should use the solution for the population of level $|2\rangle$, Eq. (34), and the relation between the longitudinal and transverse relaxation rates:

$$\gamma_{21} = \frac{1}{2T_{21}}\left(r_2^T + 1\right) + \Gamma_{21}, \tag{40}$$

where Γ_{21} is the dephasing rate at the low-frequency transition $|2\rangle - |1\rangle$ caused by elastic processes. Then we get the following expression for the anti-Hermitian part of the partial susceptibility of the probe wave:

$$\chi_p^{\mathrm{aH}} = \frac{i\eta}{4\pi|\Omega|^2}\left[\frac{\nu^2\gamma_{31}}{|\Omega|^2} + \Gamma_{21} + \frac{1}{2T_{21}}\left(1 - r_2^T\frac{T_{31}}{T_{32}}\right)\right]. \tag{41}$$

It follows from Eq. (41) that modification of the linear decrement of the probe wave in the presence of thermal excitations ($r_2^T \neq 0$) depends on the ratio between relaxation rates at two high-frequency transitions: $|3\rangle - |1\rangle$ and $|3\rangle - |2\rangle$. In a narrow range of parameters the decrement can even became negative, which corresponds to the so-called amplification without inversion regime [32]. Considering the case of close relaxation rates at high frequencies $T_{31} \approx T_{32}$, we get

$$\chi_p^{\mathrm{aH}} = \frac{i\eta}{4\pi|\Omega|^2}\left(\frac{\nu^2\gamma_{31}}{|\Omega|^2} + \Gamma_{21} + \frac{1}{2T_{21}[1+n_T(\omega_{21})]}\right). \tag{42}$$

Here we use the notation $n_T(\omega) = (e^{\hbar\omega/T} - 1)^{-1}$, which is the averaged number of "thermal" photons at frequency ω. The temperature dependence of χ_p^{aH} is defined by $n_T(\omega_{21})$ and by the temperature dependencies of the longitudinal relaxation time $T_{21}(T)$ and the "elastic" relaxation rate $\Gamma_{21}(T)$, which depend on the mechanism of dissipation that predominates under particular experimental conditions at the considered temperature. We can qualitatively model the temperature dependence of the population damping rate in the following way:

$$\frac{1}{T_{21}} = A_{21}[n_T(\omega_{21}) + 1], \tag{43}$$

which precisely corresponds to the interaction with the dissipative reservoir of harmonic oscillators (see, for example, Ref. [30]). Under real experimental conditions we should assume some dependence $A_{21}(T)$, which takes into account different dissipative mechanisms that play a major role under different temperature conditions. If Eq. (43) is used then we get that the anti-Hermitian part of the partial susceptibility of the probe wave equation (42) is not sensitive to the thermal excitations $\chi_p^{\mathrm{aH}} = \chi_p^{\mathrm{aH}}(n_T(\omega_{21}) = 0)$:

$$\chi_p^{\mathrm{aH}} = \frac{i\eta}{4\pi|\Omega|^2}\left(\frac{\nu^2\gamma_{31}}{|\Omega|^2} + \Gamma_{21} + \frac{1}{2}A_{21}\right). \tag{44}$$

It is interesting to note that the medium driven by the resonant coherent field has the negative conductivity at frequency ω_s: $-i\chi_s^{\mathrm{aH}} < 0$. The negative dissipation is caused by the process of Raman scattering of the drive wave at the transition $|1\rangle - |2\rangle$ into the Stokes satellite at frequency ω_s. The absolute value of this amplification is much less than the absorption at the frequency of the probe wave due to condition (26):

$$\left|\chi_s^{\mathrm{aH}}\right| \ll \left|\chi_p^{\mathrm{aH}}\right|. \tag{45}$$

The dependence of the anti-Hermitian component of the susceptibility at ω_s on the thermal redistribution among atomic levels is presented in Eq (38), but it is unessential due to Eq. (34). The analogous dependencies in the Hermitian parts of the susceptibilities are omitted.

So we can conclude that due to the field-induced devastation of level $|2\rangle$ [Eq. (34)] under the EIT conditions, Eq. (19), all components of the medium's susceptibility can be considered to be not sensitive to the thermal excitations of atoms in a wide range of parameters.

The radical dependence on temperature in the EIT medium appears in the noise terms of the polarization equations (37), namely, in their correlation functions.

E. Correlation functions for the noise components of the medium polarization

The commutators of noise polarization components, defined by the linear partial decrements of waves in correspondence with Eq. (15), do not depend on the thermal excitations in the medium, as follows from the conclusion of the previous section. The situation is different with correlators separately or their sum.

In linear medium the relation between correlation functions and the linear decrement is dictated by the standard fluctuation dissipation theorem [18]:

$$\begin{aligned}&\left\langle\delta\hat{P}^L_{j\nu}(z)\delta\hat{P}^{L\dagger}_{j\nu'}(z')+\delta\hat{P}^{L\dagger}_{j\nu'}(z')\delta\hat{P}^L_{j\nu}(z)\right\rangle\\&\quad=-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\chi_j^{\mathrm{aH}}(\nu)[2n_T(\omega_j+\nu)+1]\delta(\nu-\nu')\delta(z-z').\end{aligned} \tag{46}$$

We apply the technique developed in Ref. [19] to calculate the correlation functions of the noise components of polarization excited in different frequency intervals, corresponding to probe and Stokes waves, in the 4WM regime beyond the simple RWA [but using condition (26)]. Namely, we find the noise solution of Eqs. (28), and using correlation functions for the atomic noise operators, Eqs. (5) and (6), we get for correlation functions of noise components of polarizations, Eq. (36), the following expressions:

$$\left\langle\delta\hat{P}^L_{p\nu}(z)\delta\hat{P}^{L\dagger}_{p\nu'}(z')\mp\delta\hat{P}^{L\dagger}_{p\nu'}(z')\delta\hat{P}^L_{p\nu}(z)\right\rangle=-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\frac{i\eta}{4\pi|\Omega|}\left(\frac{\nu^2\gamma_{31}}{|\Omega|^3}+\frac{\gamma_{21}}{|\Omega|}\mp\frac{|\Omega|}{\gamma_{32}}\frac{n_{23}}{\rho_{11}}\right)\delta(\nu-\nu')\delta(z-z'), \tag{47}$$

$$\left\langle\delta\hat{P}^L_{s\nu}(z)\delta\hat{P}^{L\dagger}_{s\nu'}(z')\mp\delta\hat{P}^{L\dagger}_{s\nu'}(z')\delta\hat{P}^L_{s\nu}(z)\right\rangle=-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\frac{i\eta}{8\pi\omega_{21}}\left[\frac{\gamma_{31}}{2\omega_{21}}\mp\frac{2\gamma_{31}}{\omega_{21}}+\frac{2}{\omega_{21}}\left(\gamma_{31}+\frac{\gamma_{32}}{|\zeta|^2}\right)\frac{n_{23}}{\rho_{11}}\right]\delta(\nu-\nu')\delta(z-z'), \tag{48}$$

$$\left\langle\delta\hat{P}^L_{p\nu}(z)\delta\hat{P}^{L\dagger}_{s\nu'}(z')\mp\delta\hat{P}^{L\dagger}_{s\nu'}(z')\delta\hat{P}^L_{p\nu}(z)\right\rangle=0, \tag{49}$$

$$\left\langle\delta\hat{P}^L_{p\nu}(z)\delta\hat{P}^{L}_{s\nu'}(z')\mp\delta\hat{P}^{L}_{s\nu'}(z')\delta\hat{P}^L_{p\nu}(z)\right\rangle=-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\frac{i\eta e^{2i\theta}}{8\pi\omega_{21}}\left(\frac{2\gamma_{31}(\omega_{31}-\omega_p-\nu)}{|\Omega|^2}+\frac{2\gamma_{31}}{\omega_{21}}\mp\frac{2in_{23}}{\rho_{11}}\right)\delta(\nu+\nu')\delta(z-z'). \tag{50}$$

First we can see that the obtained commutators for the noise polarizations exactly coincide with the corresponding relation, Eq. (15), which guarantees the fulfillment of commutation relations for the field operators:

$$\left[\delta\hat{P}^L_{j\nu}(z)\delta\hat{P}^{L\dagger}_{j\nu'}(z')\right]=-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\chi_j^{\mathrm{aH}}(\nu)\delta(\nu-\nu')\delta(z-z'), \tag{51}$$

where $\chi_j^{\mathrm{aH}}(\nu)$ were obtained in Eq. (38). Second, the sums of correlation functions can be written in a form that resembles the standard FDT relation, Eq. (46), so that they are proportional to the anti-Hermitian components for the probe and Stokes waves, but with cardinally different proportionality factors:

$$\begin{aligned}&\left\langle\delta\hat{P}^L_{j\nu}(z)\delta\hat{P}^{L\dagger}_{j\nu'}(z')+\delta\hat{P}^{L\dagger}_{j\nu'}(z')\delta\hat{P}^L_{j\nu}(z)\right\rangle\\&\quad=\left|-i\frac{\hbar}{\pi}\frac{1}{S_\perp}\chi_j^{\mathrm{aH}}(\nu)\right|(2S_j(\nu)+1)\delta(\nu-\nu')\delta(z-z').\end{aligned} \tag{52}$$

The coefficients S_j are related to the polarization noise associated with the processes of spontaneous emission; they define the excess noise beyond that which is necessary to preserve the canonical commutation relation for the field operators. Actually the parameter S_j determines the number of spontaneously emitted photons at frequency ω_j in coherently driven medium under the condition of high optical depth.

For the probe wave this coefficient is equal to

$$S_p(\nu)=\frac{\frac{|\Omega|^2}{\gamma_{32}\gamma_{21}}\frac{n_{23}}{\rho_{11}}}{1-\frac{|\Omega|^2}{\gamma_{32}\gamma_{21}}\frac{n_{23}}{\rho_{11}}+\frac{\nu^2\gamma_{31}}{|\Omega|^2\gamma_{21}}}. \tag{53}$$

Note that it is proportional to the population of the devastated level $|2\rangle$, but with a large coefficient of proportionality $\frac{|\Omega|^2}{\gamma_{32}\gamma_{21}}$. Then, using Eq. (34) for the population ρ_{22} and Eq. (40) for the low-frequency coherence decay rate γ_{21} we get the following expression (it is written under the assumption $T_{31}\approx T_{32}$):

$$\begin{aligned}S_p(\nu)&=\frac{n_T(\omega_{21})}{\frac{1}{2}+\left(\Gamma_{21}T_{21}+\frac{\nu^2\gamma_{31}}{|\Omega|^2}T_{21}\right)[n_T(\omega_{21})+1]}\\&=\frac{n_T(\omega_{21})}{\frac{1}{2}+\frac{\Gamma_{21}}{A_{21}}+\frac{\nu^2\gamma_{31}}{|\Omega|^2A_{21}}}.\end{aligned} \tag{54}$$

In the last expression the model representation, Eq. (43), was used. It is important that the thermal noise of polarization at frequency ω_p is defined by the averaged number of thermal photons at low frequency of ground-state splitting $n_T(\omega_{21})=(e^{\hbar\omega_{21}/T}-1)^{-1}$. The number of thermal photons at splitting frequency ω_{21} may be high. Thus, for example, for Rb vapor the number of thermal photons at splitting frequency $\omega_{21} = 2\pi \times 6.83$ GHz becomes of the order of unity at temperature $T \sim 0.5$ K, while at room temperature $T = 290$ K it is already about 1000. The thermal noise factor S_p depends also on the ratio of the population exchange rate at low-frequency transition T_{21}^{-1} to the dephasing rate caused by elastic processes Γ_{21}. The thermal noise is maximum at zero detuning ν and its level increases with the ratio of population exchange rate to the rate of low-frequency coherence relaxation arising from elastic processes. In other words, the thermal noise is significant if coherence damping is mainly determined by the same mechanism of dissipation as the one that causes population transfer. The dependence of noise at frequency ω_p on the parameters of transition with frequency ω_{21} is explained by the process of parametric transfer of noise from low to high frequency in the presence of a resonant strong drive wave. It can be also interpreted as the result of spontaneous anti-Stokes Raman scattering, but modified under conditions of resonant interaction.

The analogous coefficient for the Stokes wave is

$$S_s(\nu) = 1/3. \tag{55}$$

It is interesting that it is not defined by nonzero excitations in the atomic system (thermal distribution among levels). The reason is that the fluctuations at frequency ω_s with nonzero flux can be generated due to scattering of the drive wave on zero fluctuations of the reservoir at low frequency. In other words it is caused by the spontaneous Stokes Raman scattering. For the following analysis of noise characteristics of generated two-mode squeezed radiation it was important here to find out that this coefficient is not large, so that the noise source at frequency ω_s can be ignored, since the power spectral density of the noise source at frequency ω_s is proportional to the anti-Hermitian part of the susceptibility at frequency ω_s and it is much smaller than the the noise source at frequency ω_p because Eq. (45) is fulfilled. The same concerns the cross-correlation terms in Eq. (50).

F. The solution of the coupled equations for the field operators

Using the obtained relations for the partial and the parametric components of susceptibilities, Eq. (38), we can derive the coupled equations for the filed operators according to Eq. (14). Note that these equations were derived under the condition that the nonlinear and dissipative components of polarization are small compared with the field. It is reduced to the condition imposed on the medium density:

$$\frac{\eta}{\omega_{21}} \ll 1, \tag{56}$$

where η is given by Eq. (39). So we get the following coupled equations for opposite (ν and $-\nu$) spectral components of the field operators corresponding to the probe and Stokes waves:

$$\begin{aligned}\frac{\partial \hat{a}_{p_\nu}(z)}{\partial z} - \frac{i\nu}{v_{\mathrm{gr}_p}}\hat{a}_{p_\nu}(z) + \kappa_p \hat{a}_{p_\nu}(z) &= -i\chi e^{2i\theta}\hat{a}^\dagger_{s_{-\nu}} + \hat{L}_{p_\nu},\\ \frac{\partial \hat{a}^\dagger_{s_{-\nu}}(z)}{\partial z} - \frac{i\nu}{v_{\mathrm{gr}_s}}\hat{a}^\dagger_{s_{-\nu}}(z) + \kappa_s \hat{a}^\dagger_{s_{-\nu}}(z) &= i\chi e^{-2i\theta}\hat{a}_{p_\nu} + \hat{L}^\dagger_{s_{-\nu}}.\end{aligned} \tag{57}$$

Here $\hat{a}_{j_\nu} = \sqrt{|v_{\mathrm{gr}_j}|}\hat{c}_{j_\nu}$, as defined in Sec. II B, are the operators that define the energy fluxes along the z axis in the probe and Stokes waves. The group velocities along the z axis are defined by the partial susceptibilities χ_j^H [Eq. (38)] in the following way: $v_{\mathrm{gr}_j} = c/(n_0 + 2\pi\frac{\omega_j}{n_0}\frac{\partial \chi_j^H}{\partial \nu})$, where n_0 is the refractive index of the background. So we get

$$v_{\mathrm{gr}_p} \approx \frac{2cn_0|\Omega|^2}{\eta\omega_p},$$
$$v_{\mathrm{gr}_s} \approx \frac{c}{n_0 - \frac{3\eta\omega_s}{8n_0\omega_{21}^2}}.$$

The expression for the probe-wave group velocity is presented under the condition of a strong slowdown:

$$\frac{\eta\omega_p}{|\Omega|^2} \gg 1. \tag{58}$$

It is interesting to note that under the condition $|1 - 3\eta\omega_s/8n_0^2\omega_{21}^2| < n_0^{-1}$ the group velocity of the Stokes wave exceeds the light velocity, and under the condition

$$\frac{3\eta\omega_s}{8n_0^2\omega_{21}^2} > 1 \tag{59}$$

it changes sign to negative. Such effects are well known for active and dissipative media and do not lead to violation of basic principles (see, for example, Refs. [33–35]). Note that both conditions (58) and (59) are compatible with condition (56).

The absorption coefficient of the probe wave and the amplification coefficient of the Stokes wave are given by the following relations:

$$\begin{aligned}\kappa_p &= -\frac{2\pi\omega_p}{cn_0}i\chi_p^{\mathrm{aH}} \approx \frac{\eta\omega_p}{2cn_0|\Omega|}\left(\frac{\nu^2\gamma_{31}}{|\Omega|^3} + \frac{\gamma_{21}^0}{|\Omega|}\right),\\ -\kappa_s &= \frac{2\pi\omega_s}{cn_0}i\chi_s^{\mathrm{aH}} \approx \frac{\eta\omega_s}{2cn_0\omega_{21}}\frac{3\gamma_{31}}{4\omega_{21}},\end{aligned} \tag{60}$$

where

$$\gamma_{21}^0 = \Gamma_{21} + \frac{1}{2T_{21}[1 + n_T(\omega_{21})]} = \Gamma_{21} + \frac{1}{2}A_{21}. \tag{61}$$

The Langevin noise operators $\hat{L}_{p,s_\nu} = i\sqrt{\frac{2\pi\omega_{p,s}}{\hbar cn_0}}\delta\hat{P}^L_{p,s_\nu}$. The coefficient of parametric coupling in Eq. (57) is equal to

$$\chi = \frac{\eta\sqrt{\omega_p\omega_s}}{2cn_0\omega_{21}} \approx \frac{\eta\omega_p}{2cn_0\omega_{21}}. \tag{62}$$

The solution of the system of Eqs. (57) takes the following form:

$$\begin{aligned}\begin{pmatrix}\hat{a}_{p_\nu}(z)\\ \hat{a}^\dagger_{s_{-\nu}}(z)\end{pmatrix} &= \begin{pmatrix}1\\ K_X\end{pmatrix}e^{iq_X(\nu)z}\left(\hat{u}_{X_\nu} + \int_0^z e^{-iq_X(\nu)\xi}\hat{f}_{X_\nu}d\xi\right)\\ &\quad + \begin{pmatrix}1\\ K_O\end{pmatrix}e^{iq_O(\nu)z}\left(\hat{u}_{O_\nu} + \int_0^z e^{-iq_O(\nu)\xi}\hat{f}_{O_\nu}d\xi\right),\end{aligned} \tag{63}$$

where

$$q_{O,X} = \frac{\nu}{2}\left(\frac{1}{v_{\mathrm{gr}_p}} + \frac{1}{v_{\mathrm{gr}_s}}\right) + i\frac{\kappa_p - |\kappa_s|}{2} \pm \chi\sqrt{\sigma^2 - 1},$$
$$\sigma = \frac{\nu}{2\chi}\left(\frac{1}{v_{\mathrm{gr}_p}} - \frac{1}{v_{\mathrm{gr}_s}}\right) + \frac{i}{2\chi}(\kappa_p + |\kappa_s|),$$
$$K_{O,X} = (\sigma \mp \sqrt{\sigma^2 - 1})e^{-2i\theta}. \tag{64}$$

As in Refs. [24,25], we denote two normal modes as the O mode and the X mode. The coefficients K_O and K_X define the ratio between waves with frequencies $\omega_{p,s}$ in normal modes. The amplitudes $\hat{u}_{X_\nu}$ and $\hat{u}_{O_\nu}$ are expressed via the operators of incident radiation $\hat{c}_{p,s_\nu}(0)$ defined in section $z = 0$ in vacuum using boundary condition (13):

$$\hat{u}_{X_\nu} = \sqrt{c}\frac{K_O\hat{c}_{p_\nu}(0) - \hat{c}^\dagger_{s_{-\nu}}(0)}{K_O - K_X},$$
$$\hat{u}_{O_\nu} = \sqrt{c}\frac{\hat{c}^\dagger_{s_{-\nu}}(0) - K_X\hat{c}_{p_\nu}(0)}{K_O - K_X}. \tag{65}$$

The operators $\hat{f}_{X_\nu}$ and $\hat{f}_{O_\nu}$ are the Langevin sources for the O and X modes:

$$\hat{f}_{X_\nu} = \frac{K_O\hat{L}_{p_\nu} - \hat{L}^\dagger_{s_{-\nu}}}{K_O - K_X},$$
$$\hat{f}_{O_\nu} = \frac{\hat{L}^\dagger_{s_{-\nu}} - K_X\hat{L}_{p_\nu}}{K_O - K_X}. \tag{66}$$

We take into account strong group deceleration of the probe wave and acceleration of the Stokes wave, so that $|v_{\mathrm{gr}_p}/v_{\mathrm{gr}_s}| \ll 1$, and we neglect a weak partial amplification coefficient of the Stokes wave with respect to partial absorption of the probe wave $|\kappa_s| \ll \kappa_p$ [Eq. (45)]. Then the Eqs. (64) can be simplified:

$$q_{O,X} = \chi(\sigma \pm \sqrt{\sigma^2 - 1}),$$
$$\sigma = \frac{\nu}{2\chi v_{\mathrm{gr}_p}} + \frac{i\kappa_p}{2\chi},$$
$$K_{O,X} = (\sigma \mp \sqrt{\sigma^2 - 1})e^{-2i\theta}. \tag{67}$$

Analyzing the dynamical part of solution (63) we can make the following conclusions. Under the ideal condition of no dissipation, $\kappa_p = 0$, the limitation of frequency detuning,

$$|\nu| < \Delta_P,$$

where $\Delta_P = 2\chi v_{\mathrm{gr}_p} = 2|\Omega|^2/\omega_{21}$ [see Eq. (24)], defines the band of parametric instability. If $\kappa_p = 0$ and $\nu = 0$, two ideally correlated waves are generated with equal amplitudes, so that we have

$$q_{O,X} = \pm i\chi,$$
$$K_{O,X} = \mp i e^{-2i\theta}. \tag{68}$$

Under the condition of nonzero dissipation, $\kappa_p \neq 0$, the X mode is always amplified (at every detuning) (see Refs. [25,26]), but the ratio between partial amplitudes in normal modes is changed, so that the correlations are spoiled. The regime of strong parametric instability when the solution is close to ideal, Eq. (68), is realized if the following conditions are fulfilled [see Eq. (23)]:

$$\frac{\chi}{\kappa_p} = \frac{|\Omega|^2}{\gamma^0_{21}\omega_{21}} \gg 1 \tag{69}$$

and

$$|\nu| \ll \Delta_P.$$

IV. TWO-MODE SQUEEZING

In the considered regime of parametric instability of bichromatic radiation the flow of correlated photons is generated at the output of the interaction section, and the generated radiation exhibits characteristics of two-mode squeezing. Thus the fluctuations of the sum of one quadrature of the field components and the difference of the other quadratures fall below those of the vacuum state [17]. We examine the fluctuation of the following observable:

$$\hat{X}_\nu = \frac{1}{\sqrt{2}}(\hat{X}_{p_\nu} - \hat{X}_{s_{-\nu}}),$$

where

$$\hat{X}_{j_\nu} = \hat{c}_{j_\nu}e^{i\varphi_j} + (\hat{c}_{j_\nu})^\dagger e^{-i\varphi_j}, \quad j = p,s.$$

Here the operators $\hat{c}_{j_\nu}$ are defined by the "flux" amplitudes, Eqs. (63), at the output of the layer $\hat{c}_{j_\nu} = \frac{1}{\sqrt{c}}\hat{a}_{j_\nu}(l)$. The observable $\hat{X}_\nu$ and its fluctuation may correspond to the measured spectral characteristics of the current at the detector in a balanced homodyne detection scheme, where the phases $\varphi_{p,s}$ are determined by the parameters of the local oscillator [36]. More precisely, we calculate the normalized spectral noise power defined by the averaged quadratic fluctuations of the observable $\hat{X}_\nu$ as it is related to the standard quantum limit (SQL) value. Define this quantity as

$$Q_\nu = (2\pi S_\perp c\Delta\nu_{\mathrm{det}})\overline{\langle(\Delta\hat{X}_\nu)^2\rangle}, \tag{70}$$

where the overline corresponds to the averaging over the detection resolution bandwidth $\Delta\nu_{\mathrm{det}}$. Here we take into account that

$$\overline{\langle(\Delta\hat{X}_\nu)^2\rangle}_{\mathrm{SQL}} = 1/(2\pi S_\perp c\Delta\nu_{\mathrm{det}})$$

.

A. The general solution for two-mode fluctuations

Consider the boundary condition at $z = 0$ corresponding to a completely uncorrelated state of vacuum fluctuations, so that $\langle\hat{c}_{i\nu}(0)\hat{c}^\dagger_{j\nu'}(0)\rangle = \frac{\delta(\nu-\nu')\delta_{ij}}{2\pi S_\perp c}$. With the aim to calculate the quantity Q_ν we use the obtained solutions of the equations for parametrically coupled waves, Eqs. (63), (65), and (66) with coefficient, given by Eq. (67), and the expressions for the correlation functions of the noise components of the polarizations, Eqs. (47)–(50).

As we have shown previously in Sec. III E the noise spectral density of polarization at the nonresonant frequency ω_s is small compared to that at the resonant frequency ω_p because we stay within the framework of negligible dissipation of the Stokes wave in comparison with dissipation of the probe wave, Eq. (26), and the noise associated with Raman spontaneous emission processes cannot significantly increase the level of fluctuations at frequency ω_s [see Eqs. (52) and (55)]. The analysis of the general solution for the fluctuation of the joint quadrature operator $\hat{X}_\nu$ shows that taking into account nonzero noise spectral density of polarization at frequency ω_s as well as cross-spectral density between noise polarizations at frequencies ω_s and ω_p goes beyond the approximation equation (26). So here we introduce the relation for the spectral noise power of the joint observable $\hat{X}_\nu$ under the assumption that the only source of fluctuations is the quantum and thermal noise associated with the probe field losses:

$$\begin{aligned} Q_\nu = {} & \frac{1}{|\tilde{K}_0^2-1|^2}\left[\frac{1+|K_0|^2}{2}-\left(\tilde{K}_0\frac{e^{i\Psi}}{2}+\tilde{K}_0{}^*\frac{e^{-i\Psi}}{2}\right)\right] \\ & \times\left(1+|K_0|^2-|K_0|^2\frac{\kappa_p(2S_p+1)}{\mathrm{Im}q_X}\right)e^{-2\mathrm{Im}q_X l} \\ & +\frac{1}{|\tilde{K}_0^2-1|^2}\left[\frac{1+|K_0|^2}{2}-\left(\tilde{K}_0^*\frac{e^{i\Psi}}{2}+\tilde{K}_0\frac{e^{-i\Psi}}{2}\right)\right] \\ & \times\left(1+|K_0|^2+|K_0|^2\frac{\kappa_p(2S_p+1)}{\mathrm{Im}q_X}\right)e^{-2\mathrm{Im}q_0 l} \\ & +\frac{4}{|\tilde{K}_0^2-1|^2}\mathrm{Re}\left[\left(-\mathrm{Re}\tilde{K}_0+|K_0|^2\frac{e^{i\Psi}}{2}+\frac{e^{-i\Psi}}{2}\right)\right. \\ & \left.\times\left(\mathrm{Re}\tilde{K}_0-i\frac{|K_0|^2}{|K_0|^2-1}\frac{\kappa_p(2S_p+1)}{\chi}\right)e^{i(q_X-q_0^*)l}\right]. \end{aligned} \tag{71}$$

Here $\tilde{K}_{0,X}=K_{0,X}e^{2i\theta}$ and $\Psi=2\Theta+\varphi_p+\varphi_s$. The partial absorption coefficient of the probe wave κ_p is given by Eq. (60), the coefficient of parametric coupling χ is defined by Eq. (62). The parameter defining the level of thermal fluctuations of polarization at the probe frequency S_p is given by Eq. (54). In deriving the above relation we have used that, according to Eq. (67), $\tilde{K}_0\tilde{K}_X=1$, $q_{0,X}=\chi\tilde{K}_{X,0}$, and $\mathrm{Im}q_0=-\mathrm{Im}q_X/|K_0|^2$.

The resulting relation, Eq. (71), is the sum of three terms. The first one is proportional to the growing exponent with the double increment of the X mode $|2\mathrm{Im}q_X|$. It corresponds to the exponential growing of noise associated with the instability process. The second one attenuates with the double decrement of the O mode $2\mathrm{Im}q_0$. And the third one attenuates slowly with the partial dissipation decrement $-\mathrm{Re}[i(q_X-q_0^*)]=\kappa_p$. The squeezing regime $Q_\nu\ll 1$ is realized due to appropriate phase Ψ selection that minimizes the amplitude of the first term, and the most favorable case is where the amplitude of the third term is small, but decrement of the O mode, which determines the squeezing factor, is large.

The important particular cases following from Eq. (71) are considered in what follows.

B. No dissipation

1. Center of line

In the ideal case of no dissipation in the medium, $\kappa_p=0$, at the zero detuning $\nu=0$ when Eqs. (68) are valid, we get

$$Q_\nu=\tfrac{1}{2}(1-\sin\Psi)e^{2\chi l}+\tfrac{1}{2}(1+\sin\Psi)e^{-2\chi l}.$$

Choosing $\Psi=\pi/2$ we get zero amplitude of the growing term. Due to equal amplitudes of two waves in the unstable normal mode the exponentially growing fluctuations of $X_{p,s}$ observables of two waves for the chosen phases become ideally correlated and for the difference observable these fluctuations will be completely subtracted. As the result we get

$$Q_{\nu=0}=e^{-2\chi l}. \tag{72}$$

The squeezing improves infinitely with the length of interaction.

2. Spectral characteristics

The noise spectrum under the ideal condition of no dissipation ($\kappa_p=0$) is given by the following relations obtained from Eq. (71). Within the band of parametric instability $|\nu|<\Delta_P$ we get

$$\begin{aligned} Q_\nu = {} & \frac{1}{2(1-\sigma^2)}[1-\cos(\Psi+\delta)]e^{2\chi\sqrt{1-\sigma^2}l} \\ & +\frac{1}{2(1-\sigma^2)}[1-\cos(\Psi-\delta)]e^{-2\chi\sqrt{1-\sigma^2}l} \\ & +\frac{\sigma}{1-\sigma^2}(\cos\Psi-\cos\delta), \end{aligned} \tag{73}$$

here we use the relation $|K_0|=1$; δ is defined as $\tilde{K}_0=e^{i\delta}$. For the frequency detuning larger than the width of the parametric instability band, $|\nu|>\Delta_P$ [Δ_P is defined by Eq. (24)], the noise spectrum is given by

$$\begin{aligned} Q_\nu = {} & \frac{\sigma(\sigma-\cos\Psi)}{\sigma^2-1}+\frac{1}{\sigma^2-1} \\ & \times\mathrm{Re}[(-1+\sigma\cos\Psi-2i\sqrt{\sigma^2-1}\sin\Psi)e^{-2i\chi\sqrt{\sigma^2-1}l}]. \end{aligned} \tag{74}$$

In Eqs. (73) and (74), $\sigma=\nu/\Delta_P$. It is worth noting that for every frequency detuning ν_0 within the band of parametric instability the appropriate phase Ψ can be found, namely, $\Psi=-\delta(\nu_0)$, so that the amplitude of the growing term and the amplitude of the constant term at this frequency become equal to zero and we get

$$Q_{\nu_0}=\frac{1}{1-\sigma^2}e^{-2\chi\sqrt{1-\sigma^2}l}.$$

It is obvious that the best squeezing can be achieved in the center of line $\nu=0$ and the phase $\Psi=\pi/2$. For this phase the noise level at every detuning $\nu\neq 0$ decreasing at the initial stage of interaction starts to grow exponentially beginning with the definite length. As a consequence the narrowing of the spectral interval of squeezed light takes place with an increase of the length of interaction. Analyzing Eqs. (73) and (74), we get the following estimations for the frequency band of squeezing Δ_{sq}, defined so that $Q_\nu<1$(0 dB) for $|\nu|<\Delta_{\mathrm{sq}}$. For the short distances $l\ll 1/\chi$, or more precisely for small density-length product of the medium, the weak squeezing is realized within the frequency band:

$$\Delta_{\mathrm{sq}}\approx\frac{\pi}{2}\frac{\Delta_P}{\chi l}=\pi\frac{v_{\mathrm{gr}_p}}{l}.$$

For $l=1/\chi$ the band of squeezing is equal to the parametric instability band, $\Delta_{\mathrm{sq}}=\Delta_P$. And for long distances $l\gg 1/\chi$

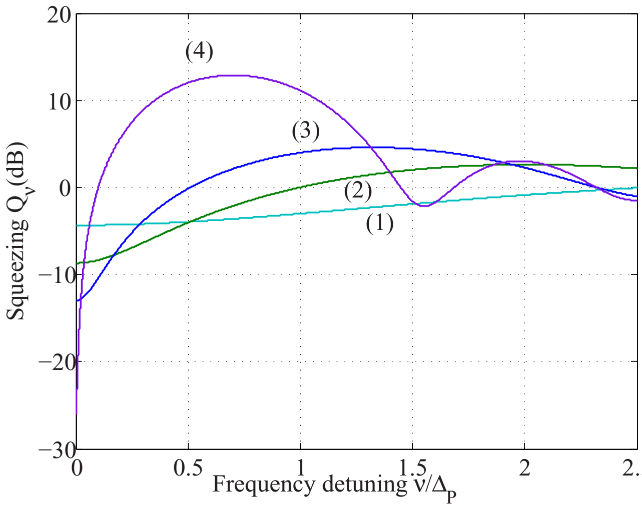

FIG. 2. Two-mode noise level Q_ν (in dB) for the phase $\Psi = \pi/2$ versus frequency detuning from the 4WM resonance ν normalized to the frequency band of the parametric instability for the case of negligible dissipation ($\kappa_p = 0$), zero temperature ($T = 0$), and different lengths of interaction: (1) $l = 0.5/\chi$, (2) $l = 1/\chi$, (3) $l = 1.5/\chi$, and (4) $l = 3/\chi$.

(large density-length product) the band of squeezing becomes much narrower,

$$\Delta_{\rm sq} \approx 2\Delta_P \exp(-\chi l), \tag{75}$$

and the magnitude of squeezing in the center becomes much higher in correspondence with Eq. (72). The effect of narrowing of the frequency band of squeezed light with the last asymptotic result, Eq. (75), was pointed in Ref. [24], where the process of generation of a biband squeezed vacuum in the medium without dissipation was analyzed in detail.

The noise spectrum Q_ν in the EIT medium without dissipation ($\kappa_p = 0$) is presented for the phase $\Psi = \pi/2$ at different lengths of interaction in Fig. 2.

C. Dissipative medium, zero temperature

How does the dissipation damage squeezing? The dissipation in the medium "delivers" an additional uncorrelated vacuum noise with amplitude, proportional to the absorption coefficient, and "spoils" correlations between two waves responsible for the squeezing. Note, that the second effect does not appear for the symmetric equations for two waves, unlike the system under consideration. Formally, nonzero partial decrement of the probe wave ($\kappa_p \neq 0$) changes the ratio between partial amplitudes in normal waves $\tilde{K}_0$ in such a way that the growing term in Eq. (71) cannot be cut off by choosing the appropriate phase Ψ any more. For the zero temperature ($S_p = 0$), at zero detuning ($\nu = 0$) and $\Psi = \pi/2$ we get from Eq. (71) the following relation:

$$\begin{aligned} Q_{\nu=0} = {} & \frac{1}{2} e^{-\kappa_p l + 2\chi\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2} l} \\ & \times \left(1 - \frac{1}{\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2}}\right)\left(1 + \frac{\kappa_p}{2\chi\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2}}\right) \\ & + \frac{1}{2} e^{-\kappa_p l - 2\chi\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2} l} \\ & \times \left(1 + \frac{1}{\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2}}\right)\left(1 - \frac{\kappa_p}{2\chi\sqrt{1+\left(\frac{\kappa_p}{2\chi}\right)^2}}\right) \\ & + e^{-\kappa_p l} \frac{\kappa_p}{2\chi\left[1+\left(\frac{\kappa_p}{2\chi}\right)^2\right]}. \end{aligned} \tag{76}$$

Even in the center of line the noise level decreases with the length of interaction only to a certain point, after which it increases exponentially. So there is an optimal length, $l_{\rm opt}$, dependent on κ_p. In the case of weak dissipation (or a strong driving field), $\kappa_p \ll \chi$ [see Eq. (69)], we get from Eq. (76)

$$l_{\rm opt} \approx \frac{1}{2\chi} \ln \frac{4\chi}{\kappa_p}. \tag{77}$$

The level of noise at this length is equal to

$$Q_{\nu=0}(l_{\rm opt}) \approx \frac{\kappa_p}{\chi} e^{-\kappa_p l_{\rm opt}} \approx \frac{\kappa_p}{\chi}\left(1 + \frac{\kappa_p}{2\chi} \ln \frac{\kappa_p}{4\chi}\right) \ll 1. \tag{78}$$

The maximal length for which $Q_{\nu=0} < 1$ is $l_{\max} = 2l_{\rm opt}$. The width of the squeezing band for the optimal length can be estimated by Eq. (75):

$$\Delta_{\rm sq} \approx \Delta_P \sqrt{\frac{\kappa_p}{\chi}} = 2|\Omega|\sqrt{\frac{\gamma_{21}^0}{\omega_{21}}}.$$

It should be noted that rather weak squeezing is realized for strong dissipation (or a weak driving field), when Eq. (69) is not fulfilled. So under the condition $\kappa_p \gg \chi$ we get for the optimal length the following relation:

$$l_{\rm opt} \approx \frac{1}{\kappa_p} \ln \frac{\kappa_p}{\chi}. \tag{79}$$

The level of noise at this length is equal to

$$Q_{\nu=0}(l_{\rm opt}) \approx 1 - \frac{2\chi}{\kappa_p},$$

and the frequency band of such squeezing is significantly higher than the frequency band of parametric instability.

Figure 3 demonstrates the interaction length dependence of squeezing in the center of line. The noise spectra for different parameters $\kappa_p(\nu = 0)/\chi$ and different lengths of interaction are shown in Fig. 4. The calculations hereinafter are made for the parameter $\frac{|\Omega|}{\omega_{21}}\sqrt{\frac{\gamma_{31}}{\gamma_{21}^0}} = 0.1$. On the one hand it provides the fulfillment of Eq. (26), and on the other hand it should be taken into account to calculate correctly the influence of the EIT resonance line shape within the considered frequency domain.

We have shown here that the main parameter that defines the level of squeezing in dissipative medium is the ratio of the partial absorption coefficient to the coefficient of parametric coupling:

$$\frac{\kappa_p}{\chi} = \frac{\gamma_{21}^0 \omega_{21}}{|\Omega|^2}.$$

The relation, analogous to Eq. (78), was obtained in Ref. [8] for the system of counterpropagating waves.

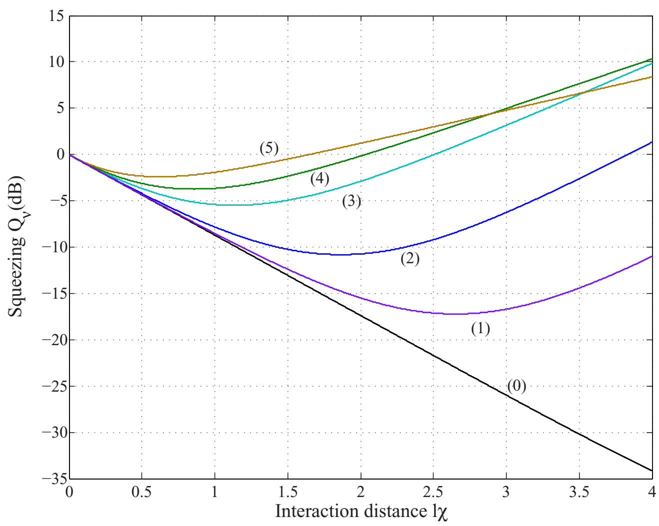

FIG. 3. Two-mode noise level Q_ν (in dB) for the phase $\Psi = \pi/2$ versus normalized length of interaction for zero detuning ($\nu = 0$), zero temperature ($T = 0$), and different parameters κ_p/χ: (0) $\kappa_p/\chi = 0$, (1) $\kappa_p/\chi = 0.02$, (2) $\kappa_p/\chi = 0.1$, (3) $\kappa_p/\chi = 0.5$, (4) $\kappa_p/\chi = 1$, and (5) $\kappa_p/\chi = 2$.

It should be noted that the existence of the optimal length of interaction dependent on the partial absorption coefficient of the probe wave was indirectly confirmed experimentally in Ref. [13], where it was shown that for a given transmission of the probe wave there is an optimal parametric gain for the best squeezing based on 4WM in an atomic vapor.

D. Thermal fluctuations squeezing

Now we analyze how the presence of thermal fluctuations in the medium may disturb generation of the squeezed state of light.

In accordance with general relation (71) the only parameter that changes the noise level of the joint quadrature operator Q_ν under the condition of thermal excitations in the medium is S_p given by Eq. (54), which in the considered nonlinear system is defined by the averaged number of thermal quanta at the low frequency of ground-state splitting $n_T(\omega_{21})$ and depends on the population damping rate and elastic dephasing rate at this transition.

Figure 5 presents the noise spectra and dependences of squeezing in the center of line on the density-length product for different values of $n_T(\omega_{21})$. The ratio A_{21}/Γ_{21} is taken equal to 1 so that $S_p(\nu = 0) = \frac{2}{3} n_T(\omega_{21})$.

Unlike the zero-temperature regime, when for every arbitrarily small parameter χ/κ_p (in frame of mentioned restrictions) the squeezing takes place, in the presence of thermal fluctuations there is a threshold value for the parameter $\chi/\kappa_p > (\chi/\kappa_p)_{\rm thr}$ dependent on S_p. The analysis of Eq. (71) for $\nu = 0$ has shown that the noise level at the optimal value for the density-length product, which depends on the parameter χ/κ_p [in the extreme cases given by Eqs. (77) and (79)], is modified in a simple way:

$$Q_{\nu=0}(l_{\rm opt}) \approx Q_{\nu=0}(l_{\rm opt})|_{S_p=0}(S_p + 1).$$

Using this relation it can be shown that in the case of low temperature, when $S_p \ll 1$, the threshold value is low, defined by the following expression:

$$\left(\frac{\chi}{\kappa_p}\right)_{\rm thr} \approx S_p/2.$$

For a strong parametric coupling regime when $\chi/\kappa_p \gg 1$, the presence of thermal fluctuations weekly changes both the peak value of squeezing and the noise spectrum.

In the case where $S_p \gtrsim 1$ the threshold value becomes essential:

$$\left(\frac{\chi}{\kappa_p}\right)_{\rm thr} \approx S_p,$$

and even under optimal conditions ($\chi \gg \kappa_p$) the noise level increases $S_p + 1$ times in comparison with the zero-temperature

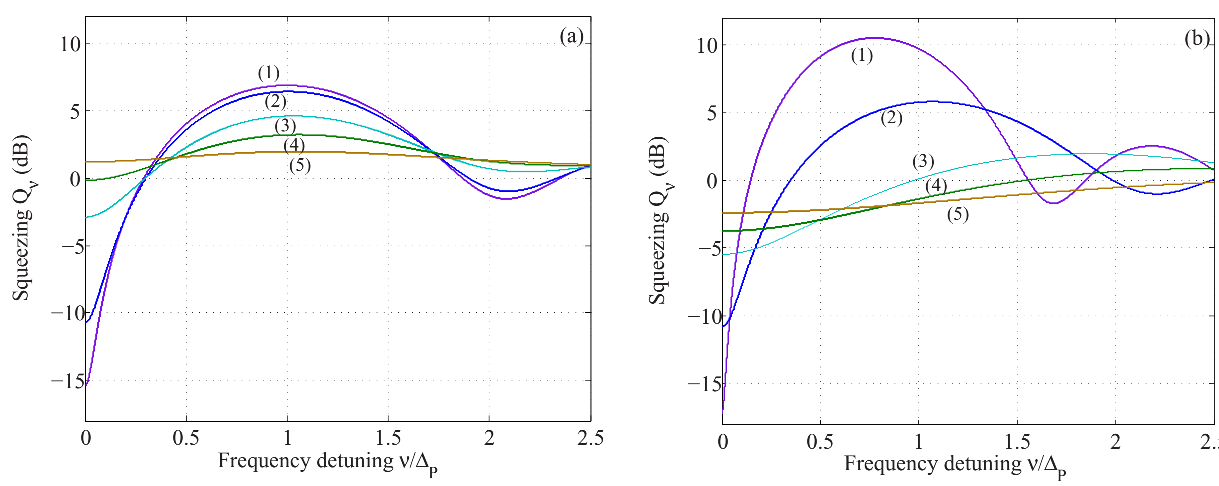

FIG. 4. Two-mode noise level Q_ν (in dB) for the phase $\Psi = \pi/2$ versus normalized detuning, for zero temperature ($T = 0$) and different parameters $\kappa_p(\nu = 0)/\chi$: (1) $\kappa_p/\chi = 0.02$, (2) $\kappa_p/\chi = 0.1$, (3) $\kappa_p/\chi = 0.5$, (4) $\kappa_p/\chi = 1$, and (5) $\kappa_p/\chi = 2$. In panel (a) all spectra are obtained at the same normalized length of interaction, $l\chi = 2$. In panel (b) each spectrum is obtained at the optimal length of interaction: (1) $l\chi = 2.65$, (2) $l\chi = 1.87$, (3) $l\chi = 1.1$, (4) $l\chi = 0.85$, and (5) $l\chi = 0.62$.

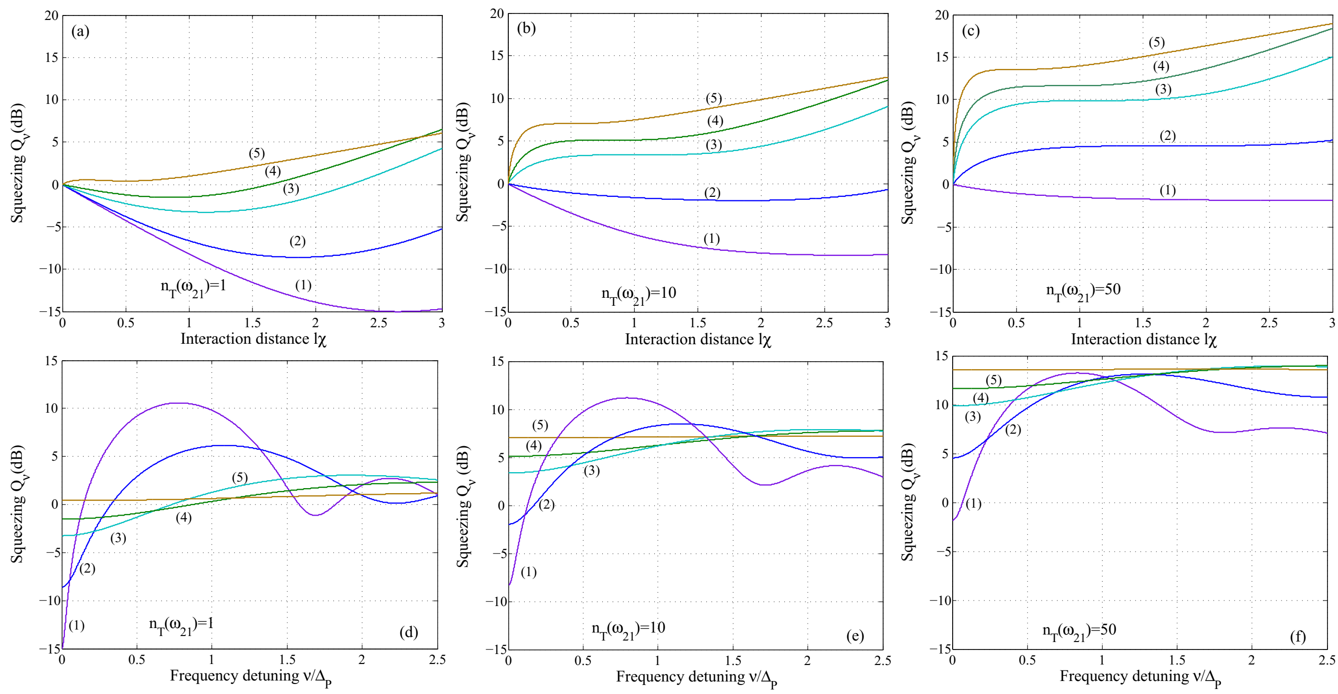

FIG. 5. Two-mode noise level Q_ν (in dB) for the phase $\Psi = \pi/2$ for different temperatures (the corresponding averaged number of thermal photons at frequency ω_{21} is pointed out in every panel) and different parameters $\kappa_p(\nu = 0)/\chi$: (1) $\kappa_p/\chi = 0.02$, (2) $\kappa_p/\chi = 0.1$, (3) $\kappa_p/\chi = 0.5$, (4) $\kappa_p/\chi = 1$, and (5) $\kappa_p/\chi = 3$. In panels (a)–(c) the length dependence of the squeezing in the center of line is presented. In panels (d)–(f) the noise spectra are presented, and each spectrum is obtained at the optimal length of interaction: (1) $l\chi = 2.65$, (2) $l\chi = 1.87$, (3) $l\chi = 1.1$, (4) $l\chi = 0.85$, and (5) $l\chi = 0.5$.

regime:

$$Q_{\nu=0}(l_{\rm opt}) \approx \frac{\kappa_p}{\chi}(S_p + 1). \quad (80)$$

An interesting result follows from Eq. (80). Namely, taking into account expressions for the absorption coefficient κ_p [Eqs. (60) and (61)] and for the parameter S_p [Eq. (54)], we get that Eq. (80) can be presented in the following very simple form:

$$Q_{\nu=0}(l_{\rm opt}) \approx \frac{\gamma_{21}(T)\omega_{21}}{|\Omega|^2}. \quad (81)$$

It is important to note that this relation takes into account both the modification of the relation for the absorption coefficient in the presence of thermal excitations in the medium and the additional noise associated with the spontaneous Raman emission that appears in the presence of thermal excitations. However, the resulting expression for the level of noise of the squeezed two-mode quadrature $\hat{X}_\nu$ under optimal conditions ($\chi/\kappa_p \gg 1$, $l = l_{\rm opt}$, $\nu = 0$) is so constructed that it can be calculated neglecting both effects. The only temperature-dependent parameter in the resulting expression is the low-frequency coherence damping rate γ_{21} that characterizes the atomic system at the actual temperature. Therefore we can conclude that under optimal conditions the thermal excitations in the atomic system affect the level of two-mode squeezing only inasmuch as they increase the damping rate of low-frequency coherence.

V. DISCUSSION

We presented here the detailed analytical investigation of two-mode squeezed-vacuum generation in a robust scheme of four-wave mixing in a resonant Λ configuration taking into account such negative factors as dissipation caused by relaxation in the atomic system and thermal excitations delivering the additional uncorrelated noise. These processes are considered on the basis of a self-consistent microscopic approach. We investigated the influence of spontaneous Raman scattering under resonant conditions of EIT on the level of two-mode squeezing. We obtained the analytical formulas for the optimal density-length product of atomic medium and for the frequency width of the squeezing band as they depend on the drive intensity and the relaxation rate.

The following illustrative estimations of real experimental parameters, for example, for ^{87}Rb vapor ($D1$ transition; wavelength of resonant radiation, $\lambda = 795$ nm; frequency of ground-state splitting, $\omega_{21} = 6.83$ GHz) can be made. For the relaxation rate $\gamma_{21} = 15$ kHz, if the drive power is about 10 mW and the beam focusing diameter is 2 mm (so that the Rabi frequency $|\Omega| \approx 30$ MHz) the condition of strong parametric instability, Eq. (69), is fulfilled: $\chi/\kappa_p \approx 8$. Then the calculated dimensionless optimal density-length product $l_{\rm opt}\chi = 1.7$ corresponds to the following dimension value: $lN \approx 4$ cm $\times\, 7 \times 10^{11}$ cm^{-3}. The level of squeezing in the center of line is equal to -10 dB and the frequency band of squeezing $\Delta_{\rm sq} \approx 80$ kHz. Note that here we used rather moderate relaxation parameters. The minimization of the ground-state coherence damping rate is an "old problem," which has been effectively solved in relation to the development of CPT-based atomic frequency standards (see, for example, Ref. [37]). Damping rates of the order of 10 Hz are achievable nowadays at room temperatures. Therefore we can state that the particular problem of thermal noise influence on the squeezing can potentially be solved to the same extent.

So the following general conclusion can be made. Strong parametric coupling in a regime of four-wave mixing in a Λ scheme of three-level atoms may provide two-mode squeezing not only of the intrinsic quantum fluctuations of light but also squeezing of thermal fluctuations. The highest attainable level of squeezing taking into account squeezing of thermal noise is dictated by the ground-state coherence relaxation rate and the intensity of the drive field.

ACKNOWLEDGMENTS

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APPENDIX: CORRELATION FUNCTIONS OF ATOMIC LANGEVIN OPERATORS

Here we analyze the correlation properties of Langevin operators in the atomic equation, Eq. (2):

$$\dot{\hat{\rho}}_{mn} = -\frac{i}{\hbar}(\hat{h}_{mp}\hat{\rho}_{pn} - \hat{\rho}_{mp}\hat{h}_{pn}) + \hat{R}_{mn} + \hat{F}_{mn}. \quad \text{(A1)}$$

The important point is that the properties of the noise operators $\hat{F}_{mn}$ are connected with the properties of the relaxation operators $\hat{R}_{mn}$ [38].

The model of constant relaxation rates, Eq. (3), obtained within the frame of the Markov approximation [38] is equivalent to the δ correlation in time of the noise source:

$$\langle \hat{F}_{mn}(\mathbf{r},t)\hat{F}_{pq}(\mathbf{r}',t')\rangle = 2D_{mnpq}(\mathbf{r},\mathbf{r}',t)\delta(t-t'). \quad \text{(A2)}$$

This approximation allows one to use so-called generalized Einstein relations [31,39] for calculating the diffusion coefficients $D_{mnpq}(\mathbf{r},\mathbf{r}',t)$:

$$\begin{aligned}2D_{mnpg}(\mathbf{r},\mathbf{r}') &= \frac{d}{dt}\langle\hat{\rho}_{mn}(\mathbf{r},t)\hat{\rho}_{pq}(\mathbf{r}',t)\rangle\\ &\quad -\left\langle\left(\frac{d}{dt}\hat{\rho}_{mn}(\mathbf{r},t)-\hat{F}_{mn}(\mathbf{r},t)\right)\hat{\rho}_{pq}(\mathbf{r}',t)\right\rangle\\ &\quad -\left\langle\hat{\rho}_{mn}(\mathbf{r},t)\left(\frac{d}{dt}\hat{\rho}_{pq}(\mathbf{r}',t)-\hat{F}_{pq}(\mathbf{r}',t)\right)\right\rangle.\end{aligned} \quad \text{(A3)}$$

Next, we assume that the action of the reservoir on different atoms is independent, so that fluctuations of the density matrix operators for different atoms are not correlated: $\langle(\hat{\rho}_{mn;j} - \langle\hat{\rho}_{mn;j}\rangle)(\hat{\rho}_{pq;i} - \langle\hat{\rho}_{pq;i}\rangle)\rangle \propto \delta_{ij}$. Taking into account also the strict equality that should be fulfilled for each atom by definition of the density matrix operators, $\hat{\rho}_{mn;j}\hat{\rho}_{pq;j} = \hat{\rho}_{pn;j}\delta_{mq}$, we get for the averaged product of the space-dependent density matrix operators the following relation:

$$\langle\hat{\rho}_{mn}(\mathbf{r})\hat{\rho}_{pq}(\mathbf{r}')\rangle = \langle\hat{\rho}_{mn}(\mathbf{r})\rangle\langle\hat{\rho}_{pq}(\mathbf{r}')\rangle + \delta_{mq}\langle\hat{\rho}_{pn}(\mathbf{r})\rangle\delta(\mathbf{r}-\mathbf{r}'). \quad \text{(A4)}$$

The expression for the diffusion coefficients, Eq. (A3), with regard to Eqs. (A4), (2), and (3), finally takes δ correlated in space form:

$$D_{mnpq}(\mathbf{r},\mathbf{r}',t) = D_{mnpq}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}'), \quad \text{(A5)}$$

where

$$2D_{mnpq}(\mathbf{r},t) = \delta_{mq}\langle\hat{R}_{pn}\rangle - \sum_l r_{mnql}\langle\hat{\rho}_{pl}\rangle - \sum_l r_{pqlm}\langle\hat{\rho}_{ln}\rangle.$$

In a simple case of Eqs. (4), the correlation functions of the Langevin sources for the "off-diagonal" operators ($m \neq n$, $p \neq q$) are given by the following expression:

$$2D_{mnpq}(\mathbf{r},t) = \delta_{mq}((\gamma_{mn}+\gamma_{pq})\langle\hat{\rho}_{pn}\rangle + \langle\hat{R}_{pn}\rangle). \quad \text{(A6)}$$

Thus, the autocorrelation function for Langevin operator at some transition $m-n$ is given by

$$2D_{mnnm}(\mathbf{r},t) = 2\gamma_{mn}\langle\hat{\rho}_{nn}\rangle + \langle\hat{R}_{nn}\rangle. \quad \text{(A7)}$$

The excited coherence at some transition $|a\rangle - |b\rangle$ corresponds to the nonzero correlations of Langevin sources at the adjacent atomic transitions:

$$2D_{mabm}(\mathbf{r},t) = (\gamma_{am}+\gamma_{bm}-\gamma_{ab})\langle\hat{\rho}_{ba}\rangle. \quad \text{(A8)}$$

For the spectral components of the Langevin operators defined as $\hat{F}_{mn}(\mathbf{r},t) = \int_{-\infty}^{+\infty}\hat{F}_{mn}(\mathbf{r},\omega)e^{-i\omega t}d\omega$, taking into account Eqs. (A2) and (A5), we get

$$\langle\hat{F}_{mn}(\mathbf{r},\omega)\hat{F}_{pq}(\mathbf{r}',\omega')\rangle = \frac{1}{\pi}D_{mnpq}(\mathbf{r},\omega+\omega')\delta(\mathbf{r}-\mathbf{r}'), \quad \text{(A9)}$$

where

$$D_{mnpq}(\mathbf{r},\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty}D_{mnpq}(\mathbf{r},t)e^{i\omega t}dt. \quad \text{(A10)}$$

Under the adiabatic approximation, neglecting the slow evolution of populations and the amplitude of drive-induced coherence in the resonant approximation, $\langle\hat{\rho}_{nn}\rangle \approx$ const, $\langle\hat{\rho}_{ba}\rangle = \sigma_{ba}e^{\mp i\omega_d t}|_{b\gtrless a}$, $\sigma_{ba} \approx$ const, the atomic noise operators are δ correlated in frequency, and we get from Eqs. (A7) and (A8) the following correlation functions:

$$\begin{aligned}&\langle\hat{F}_{mn}(\mathbf{r},\omega)\hat{F}_{nm}(\mathbf{r}',\omega')\rangle\\ &\quad = \frac{1}{2\pi}(2\gamma_{mn}\langle\hat{\rho}_{nn}\rangle + \langle\hat{R}_{nn}\rangle)\delta(\omega+\omega')\delta(\mathbf{r}-\mathbf{r}'),\end{aligned} \quad \text{(A11)}$$

$$\begin{aligned}&\langle\hat{F}_{ma}(\mathbf{r},\omega)\hat{F}_{bm}(\mathbf{r}',\omega')\rangle\\ &\quad = \frac{1}{2\pi}(\gamma_{am}+\gamma_{bm}-\gamma_{ab})\sigma_{ba}\delta(\omega+\omega'\mp\omega_d)|_{b\gtrless a}\delta(\mathbf{r}-\mathbf{r}').\end{aligned} \quad \text{(A12)}$$

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