Light-intensity ssusceptibility and "active" noise spectroscopy

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In this paper, we consider informative potentialities of the "active" optical noise spectroscopy, under which we understand, generally, spectroscopy of response of a multilevel quantum system to the resonant optical field with its intensity modulated by "white" noise. We show that calculations of such a response can be most conveniently performed, in the linear approximation, by introducing the notion of light-intensity susceptibility (LIS) whose spectrum is determined by Laplace transform of the response to a small step-wise change of the optical field intensity. The results of calculations for a simple four-level quantum system show that its LIS spectrum may provide information not only about the ground-state structure (like conventional Faraday-rotation-based spin noise spectroscopy), but also about properties of the optical transitions (including nutation frequencies in the applied optical field). From the experimental point of view, such a noise spectroscopy of the intensity-related susceptibility can be especially efficient in combination with the up-to-date spectrum analyzers providing extremely fast data processing.

I. INTRODUCTION

Optical spectroscopy, with its frequencies lying in the range of 10^{15} Hz, is known to be capable of studying spectral features of the system at frequencies many orders of magnitude lower (like, e.g., Zeeman or hyperfine splittings). Such a high-resolution spectroscopy often employs, for detection of these features, the intensity correlation characteristics of the light coupled to the system. Ideology of the light intensity noise spectroscopy has been primarily realized by Forrester [1] and Hunbury-Brown and Twiss [2] and then has been further developed for studying dynamics of motion of different atomic and molecular systems including dynamics of nonstationary quantum states (see, e.g., [3]). With the advent of lasers, the light intensity noise spectroscopy has crucially increased its sensitivity and gained a deeper practical sense.

The methods of noise spectroscopy intended for studying spectra of eigen-frequencies of quantum systems can be divided into two main classes. In the measurements of the first class, optical probing of the sample is supposed to be nonperturbative: the light beam passing through the sample in the region of its transparency acquires additional intensity or polarization fluctuations that carry the sought information. Ideally, intrinsic noise of the probe beam, in this approach, is negligibly small. A typical example of the experimental technique of this class is the Faraday-rotation-based spin noise spectroscopy (SNS), which provides a unique opportunity of studying magnetic resonance and spin dynamics of atomic and solidstate systems practically in a perturbation-free way [4–7]. The recent enormous growth of interest to this experimental technique, primarily demonstrated more than 30 years ago [4], is associated, to a considerable extent, with great progress in the present-day electronics, which has made it possible to digitize arrays of data with the sampling rates up to several GHz and to perform fast Fourier transform (FFT) in real time. As a result, the new class of spectrum analyzers with FFT has allowed to increase sensitivity of the measurements and, correspondingly, to shorten the acquizition time by several orders of magnitude (as compared to the standard sweeping spectral analyzers). Due to this technical refinement, SNS has been successfully applied to semiconductor systems, including microsamples and nanostructures [6] and is now about to turn into a routine tool of magnetic spectroscopy.

The other experimental approach to the noise spectroscopy that has been developed in 90s and applied to atomic systems, is based on the use of a randomly modulated light beam acting on the system in the region of its absorption [8–13]. This technique is deliberately perturbative: the system excited by the pump light modulated in a wide frequency range responds selectively to certain resonant frequencies, and, as a result, the noise spectrum of the transmitted light appears to be modified. In the above papers, as a source of light was used a diode laser with specific correlation properties of its output emission: intensity fluctuations were extremely low (they were claimed to lie below the shot-noise level [14]), whereas its frequency noise was well pronounced. The frequency modulated light, due to interaction with a narrow spectral line of the atomic medium, produced the required noisy excitation of the system. It was shown, in those experiments that the atomic system, under certain conditions, may exhibit resonant response not only at the frequency of the ground-state magnetic resonance (Larmor precession), as it was primarily demonstrated by Bell and Blum[15], but also at Rabi frequency of the resonant transition[10], which may substantially widen informative potential of the noise spectroscopy.

It is noteworthy that this kind of noise spectroscopy was never considered as a possible experimental tool for studying a wider circle of materials, including semiconductor systems and nanostructures, highly important for

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contemporary physics and applications. At the same time, from the methodological viewpoint, experimental measurements of the intensity-related susceptibility using fluctuating optical fields have much in common with the technique of conventional "passive" noise spectroscopy and thereby have all chances to take an advantage of the new instrumental opportunities provided by the upto-date electronics.

In this paper, we consider a general theoretical model that describes the effect of a four-level system on the intensity noise spectrum of the transmitted resonant light. The light intensity noise spectrum is assumed to be "white" and essentially exceeding in magnitude the shotnoise level. We analyze interaction of the light beam with the medium by introducing the notion of the light-intensity susceptibility (LIS), which in our opinion, may be useful for solving certain problems of nonlinear optics. In particular, this notion was, in fact, used in the analysis of dynamics of a saturable absorber in modulated optical fields [16].

In what follows we study spectral behavior of the intensity-related susceptibility in the range of relatively low frequencies (e.g., of the ESR range). We will show that frequency dependence of the light-intensity susceptibility contains information not only about energy and relaxation characteristics of the system accessible for linear spectroscopy, (including conventional "passive" spin noise spectroscopy), but also about specific dynamics of optical transitions (nutation frequencies) in the field of the noisy resonant pumping. We will see that, in contrast to the conventional linear optical spectroscopy, which allows one to measure frequencies of transitions between the levels of unperturbed states of the system, the LIS spectroscopy makes it possible (for sufficiently long relaxation times) to measure frequencies of transitions between the states of some effective Hamiltonian dependent on the optical field intensity I_0 .[17]

II. STARTING POINTS

We define the light-intensity susceptibility (LIS) in the following way. Assume that the system under study is subjected to action of a quasi-monochromatic optical field of the frequency ω , with its intensity I having a dc (I_0) and weak ac $(\delta I(t))$ components $I=I_0+\delta I(t)$. Correspondingly, the power P absorbed by the system will also have the dc and ac components: $P=P_0+\delta P(t)$. We define the light-intensity susceptibility as a linear susceptibility $\chi(\imath\xi)$ connecting the ac components of the intensity and absorption in the case when $\delta I(t) \to \delta I e^{-\imath \xi t}, \delta P(t) \to \delta P e^{-\imath \xi t}$ and $\delta P=\chi(\imath\xi)\delta I$.

A possible experimental observation of frequency dependence (spectrum) of the light-intensity susceptibility with the use of the spectrum analyzer looks as follows [Fig. 1(a)]. The laser beam subjected to a broadband noise intensity modulation [18] passes through the sample under study and hits the photodetector, whose out-

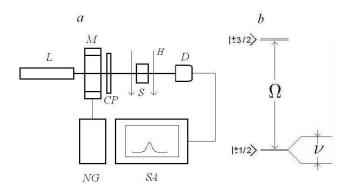


Figure 1. (a) schematic of experimental setup for observation of light intensity susceptibility. L – laser, M – optical modulator, CP – circular polarizer, S – sample, D – photodetector, SA – spectrum analyzer, NG – noise generator. (b) energy structure of the model system.

put signal is fed to the input of the spectrum analyzer. If the intensity modulation spectrum of the laser beam is "white", then we will observe, at the output of the spectrum analyzer, the LIS spectrum of the studied sample. The experiments of this kind have been described in [8, 11] where the resonant paramagnetic susceptibility of atomic gases has been detected. It should be emphasized that, though we consider a linear intensity susceptibility, it can be revealed only by optically nonlinear media. In the next section, we will calculate the LIS of the simplest model system and will study its spectrum.

III. MODEL CALCULATIONS

The sample under study is assumed to be an ensemble of particles, and the total power absorbed by the sample irradiated by the electromagnetic field to be equal to the sum of contributions of individual particles. The energy spectrum of individual particle is assumed to consist of two doublets [Fig. 1(b)]: the ground $|\pm 1/2\rangle$ and the excited $|\pm 3/2\rangle$ one (here we indicate projection of the angular moment onto the light propagation direction, which is assumed to coincide with the z-axis of the our coordinate system). This energy structure arises, in particular, in the atomic multiplet perturbed by an axially symmetric field. [19] In this case, magnetic splitting of the ground doublet is characterized by isotropic q-factor, whereas the doublet $|\pm 3/2\rangle$ is split only by the field parallel to the z-axis. Spectral distance between the doublets (denoted by Ω) is supposed to be close to frequency ω of the optical field $\omega \approx \Omega$ and essentially higher than other frequency parameters of the problem.

We will calculate the LIS spectrum we are interesting in as follows. Let us assume that the system under study was subjected, during sufficiently long time, to a monochromatic optical field with fixed intensity, so that the power absorbed by the system has already reached

its steady-state value. Let the amplitude of this field now experience a jump that induces a transient process corresponding to transition to a new stationary state. Below, we will find the transient characteristics (i.e., temporal behavior of the absorbed power during this process), which is connected with the sought susceptibility by the Laplace transform.

Let us pass to implementation of the above program for the conditions of the experiment [15]. The optical field acting upon the system is assumed to be circularly polarized and the whole system to be placed in a transverse magnetic field \mathcal{H} directed along the x-axis [Fig. 1(a)].

Since the magnetic splitting of the ground doublet is supposed to be small compared with the optical frequency ω , among the matrix elements of the operator of interaction of the system with optical field (denote it H_1), we have to take into account only those corresponding to quasi-resonant transitions:

$$\begin{cases} \langle -1/2|H_1| + 1/2 \rangle = \text{ non resonant} \\ \langle +1/2|H_1| - 1/2 \rangle = \text{ non resonant} \\ \langle +3/2|H_1| + 1/2 \rangle = Ae^{-\imath \omega t} \\ \langle -3/2|H_1| - 1/2 \rangle = Ae^{\imath \omega t} \\ \langle +1/2|H_1| + 3/2 \rangle = Ae^{\imath \omega t} \\ \langle -1/2|H_1| - 3/2 \rangle = Ae^{-\imath \omega t} \end{cases}$$

Here, the constant A describes intensity of the transitions under circularly polarized excitation. If we arrange possible states of the system in the order $|+1/2\rangle$, $|-1/2\rangle$, $|-3/2\rangle$, then the matrix H_1 will have the form

$$H_1 = A \begin{pmatrix} 0 & 0 & 0 & \mu \\ 0 & 0 & \bar{\mu} & 0 \\ 0 & \mu & 0 & 0 \\ \bar{\mu} & 0 & 0 & 0 \end{pmatrix} \qquad \mu \equiv e^{i\omega t} \tag{1}$$

At A = 0, the Hamiltonian matrix of the system, in the basis introduced above, will have the form

$$H_0 = \begin{pmatrix} 0 & \nu & 0 & 0 \\ \nu & 0 & 0 & 0 \\ 0 & 0 & \Omega & 0 \\ 0 & 0 & 0 & \Omega \end{pmatrix} \qquad \nu \equiv \frac{g\beta\mathcal{H}}{2\hbar} \tag{2}$$

At $A \neq 0$, the Hamiltonian of the system depends on time and equals $H = H_0 + H_1$. Neglecting the relaxation processes (they will be taken into account later), dynamics of the density matrix of the system ρ can be described by the Liouville equation $i\dot{\rho} = [H,\rho]$. To treat this equation, it is convenient to pass to the basis, in which H_0 is diagonal, and then to the "rotating coordinate frame", where the total Hamiltonian is time-independent. The first step is made using a unitary transformation with the matrix

$$S \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 & 0\\ 1 & -1 & 0 & 0\\ 0 & 0 & \sqrt{2} & 0\\ 0 & 0 & 0 & \sqrt{2} \end{pmatrix}, \text{ with } S^{-1} = S \quad (3)$$

We will mark with tilde all the operators having the form $\tilde{H} \equiv SHS$. Then, direct calculations show that

$$\tilde{H}_0 = \begin{pmatrix} \nu & 0 & 0 & 0 \\ 0 & -\nu & 0 & 0 \\ 0 & 0 & \Omega & 0 \\ 0 & 0 & 0 & \Omega \end{pmatrix} \tag{4}$$

$$\tilde{H}_{1} = \frac{A}{\sqrt{2}} \begin{pmatrix} 0 & 0 & \bar{\mu} & \mu \\ 0 & 0 & -\bar{\mu} & \mu \\ \mu & -\mu & 0 & 0 \\ \bar{\mu} & \bar{\mu} & 0 & 0 \end{pmatrix}$$
 (5)

And the density matrix $\tilde{\rho} = S\rho S$ meets the equation $i\partial\tilde{\rho}/\partial t = [\tilde{H},\tilde{\rho}]$. Transition to the "rotating coordinate frame" is performed using the following time-dependent transformation of the operators. Let us pass to the hatted operators and new density matrix using the rule

$$\hat{H} = e^{iMt} \tilde{H} e^{-iMt} \quad \sigma \equiv e^{iMt} \tilde{\rho} e^{-iMt}$$
 (6)

Where the matrix M has the following elements

Then, one can easily see that σ satisfies the equation $i\dot{\sigma}=[\widehat{H}-M,\sigma]$. Calculations of the matrix \widehat{H} show that it contains time-independent elements and elements proportional to $e^{\pm 2\imath\omega t}$. The matrix elements oscillating at double frequency are ignored as essentially nonresonant and not affecting dynamics of the density matrix σ . Thus, the matrix \widehat{H} , with allowance for the aforesaid, can be written in the form:

$$\widehat{H} = \begin{pmatrix} \nu & 0 & 0 & d \\ 0 & -\nu & 0 & d \\ 0 & 0 & \Omega & 0 \\ d & d & 0 & \Omega \end{pmatrix} \qquad d \equiv \frac{A}{\sqrt{2}} \tag{8}$$

It follows from this expression that the third column and third row of the matrix H are mutually diagonal and can be omitted. Now, all the operators can be presented by 3×3 matrices, and the equation of motion for the matrix σ acquires the form

$$i\dot{\sigma} = [W, \sigma], \qquad W \equiv \begin{pmatrix} \nu & 0 & d \\ 0 & -\nu & d \\ d & d & \Delta \end{pmatrix}, \qquad \Delta \equiv \Omega - \omega$$
(9)

with the time-independent matrix W.

As mentioned above, our task is to calculate transient dynamics of the power P absorbed by the system after a jump of the optical field amplitude. This power can be expressed in the form

$$P = \frac{d}{dt} \operatorname{Sp} \rho H = \frac{d}{dt} \operatorname{Sp} \sigma \widehat{H} = \operatorname{Sp} \dot{\sigma} \widehat{H} + \operatorname{Sp} \sigma \frac{d\widehat{H}}{dt}$$
(10)

The last term in (10) can be neglected because the operator \widehat{H} oscillates in time at double optical frequency, and the mean value of the corresponding contribution vanishes. Thus,

$$P = \operatorname{Sp} \dot{\sigma} \widehat{H} = -i \operatorname{Sp} [\widehat{H} - M, \sigma] \widehat{H} = i \operatorname{Sp} [M, \sigma] \widehat{H}$$
(11)

Using explicit formulas (8) and (9) for the matrices Mand \widehat{H} , we finally have

$$P = 2\omega d \left[\text{Im } \sigma_{13} + \text{Im } \sigma_{23} \right] \tag{12}$$

Note that account of relaxation terms in the equation of motion for the density matrix leads to appearance of terms proportional to the relaxation rates in the expression for the absorbed power P. We will assume that these rates are much lower than the optical frequency ω entering Eq (12) and will use Eq. (12) in the presence of relaxation processes also.

Now, we will write down Eq. (9) in components of the density matrix and introduce relaxation needed to obtain the steady-state regime of absorption:

$$\begin{cases}
i\dot{\sigma}_{12} = \sigma_{12}[2\nu - iT_{12}^{-1}] + d[\sigma_{32} - \sigma_{13}] \\
i\dot{\sigma}_{21} = -\sigma_{21}[2\nu + iT_{21}^{-1}] + d[\sigma_{31} - \sigma_{23}] \\
i\dot{\sigma}_{13} = \sigma_{13}[\nu - \Delta - iT_{13}^{-1}] + d[\sigma_{33} - \sigma_{11} - \sigma_{12}] \\
i\dot{\sigma}_{31} = \sigma_{31}[\Delta - \nu + iT_{31}^{-1}] + d[\sigma_{11} + \sigma_{21} - \sigma_{33}] \\
i\dot{\sigma}_{23} = \sigma_{23}[-\Delta - \nu - iT_{23}^{-1}] + d[\sigma_{33} - \sigma_{21} - \sigma_{22}] \\
i\dot{\sigma}_{32} = \sigma_{32}[\Delta + \nu + iT_{32}^{-1}] + d[\sigma_{22} - \sigma_{33} + \sigma_{12}] \\
i\dot{\sigma}_{11} = d[\sigma_{31} - \sigma_{13}] + \sigma_{33}\tau_{1}^{-1} + (\sigma_{22} - \sigma_{11})T_{1}^{-1} \\
i\dot{\sigma}_{22} = d[\sigma_{32} - \sigma_{23}] + \sigma_{33}\tau_{1}^{-1} - (\sigma_{22} - \sigma_{11})T_{1}^{-1} \\
i\dot{\sigma}_{33} = d[\sigma_{13} - \sigma_{31} + \sigma_{23} - \sigma_{32}] - 2\sigma_{33}\tau_{1}^{-1}
\end{cases}$$
(13)

The form of the relaxation terms for diagonal elements of the density matrix implies that states 1 and 2 are the lowest, and the temperature is so high that they are equally populated. State 3, on the contrary, is assumed to lie

so high that its equilibrium population is zero. These assumptions correspond to the conditions $g\beta\mathcal{H} \ll kT$ and $\hbar\Omega >> kT$. Let us introduce the following notations for the real and imaginary parts of the matrix elements σ and relaxation times

$$\begin{cases}
\sigma_{12} + \sigma_{21} = x_1 \\
\sigma_{13} + \sigma_{31} = x_2 \\
\sigma_{23} + \sigma_{32} = x_3
\end{cases}$$
(14a)

$$\begin{cases} \sigma_{12} - \sigma_{21} = iy_1 \\ \sigma_{13} - \sigma_{31} = iy_2 \\ \sigma_{23} - \sigma_{32} = iy_3 \end{cases}$$
 (14b)

$$\begin{cases}
\sigma_{11} = s_1 \\
\sigma_{22} = s_2 \\
\sigma_{33} = s_3
\end{cases}$$
(14c)

$$\begin{cases}
\sigma_{12} + \sigma_{21} = x_1 \\
\sigma_{13} + \sigma_{31} = x_2 \\
\sigma_{23} + \sigma_{32} = x_3
\end{cases}$$

$$\begin{cases}
\sigma_{12} - \sigma_{21} = iy_1 \\
\sigma_{13} - \sigma_{31} = iy_2 \\
\sigma_{23} - \sigma_{32} = iy_3
\end{cases}$$

$$\begin{cases}
\sigma_{11} = s_1 \\
\sigma_{22} = s_2 \\
\sigma_{33} = s_3
\end{cases}$$

$$\begin{cases}
T_{12} = T_{21} = T_2 \\
T_{13} = T_{31} = \tau_2 \\
T_{23} = T_{32} = \tau_2
\end{cases}$$
(14a)

Here, we denoted the dephasing time in the magnetic doublet as T_2 and the dephasing time in the transitions 2-3 and 1-3, saturated by the optical field, as τ_2 . By defining the vector-column v as

$$v = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ y_1 \\ y_2 \\ y_3 \\ s_1 \\ s_2 \\ s_3 \end{pmatrix}$$
 (15)

and the matrix G(d) as

$$G(d) = \begin{pmatrix} -T_2^{-1} & 0 & 0 & 2\nu & -d & -d & 0 & 0 & 0\\ 0 & -\tau_2^{-1} & 0 & -d & -\Delta + \nu & 0 & 0 & 0 & 0\\ 0 & 0 & -\tau_2^{-1} & d & 0 & -\Delta - \nu & 0 & 0 & 0\\ -2\nu & d & -d & -T_2^{-1} & 0 & 0 & 0 & 0 & 0\\ d & \Delta - \nu & 0 & 0 & -\tau_2^{-1} & 0 & 2d & 0 & -2d\\ d & 0 & \Delta + \nu & 0 & 0 & -\tau_2^{-1} & 0 & 2d & -2d\\ 0 & 0 & 0 & 0 & -d & 0 & -T_1^{-1} & T_1^{-1} & \tau_1^{-1}\\ 0 & 0 & 0 & 0 & 0 & -d & T_1^{-1} & -T_1^{-1} & \tau_1^{-1}\\ 0 & 0 & 0 & 0 & d & d & 0 & 0 & -2\tau_1^{-1} \end{pmatrix},$$
(16)

Using Eq.(13), we obtain the following equation for v

$$\frac{dv}{dt} = G(d)v\tag{17}$$

Since the last row of matrix G is the sum of two previous ones, this matrix should always have a zero eigennumber. The corresponding eigen-vector (denote it e^9) allows us to obtain the steady-state regime of the system saturated by the optical field. The vector b, corresponding to the steady-state matrix of the system, evidently represents the vector e^9 normalized as $b = q^{-1}e^9$, where $q \equiv e_7^9 + e_8^9 + e_9^9$. In accordance with the formulation of the problem, given above, we assume that the density matrix of the system, before the jump of the optical field amplitude, was determined by the vector b. In this case, the power P_0 absorbed by the system before the jump can be expressed through the components of this vector using Eq. (12)

$$P_0 = \omega d[b_5 + b_6] \tag{18}$$

If the jump of the optical field amplitude $d \to d + \delta d$ occurs at t=0, then, to find dynamics of the system after the jump, we have to solve the equation $\dot{v}=G(d+\delta d)v$ with the initial condition v(t=0)=b. Denote by $p^i, i=1,2,...,9$ the eigen-vectors of the matrix $G(d+\delta d)$ and decompose the vector b over them:

$$b = \sum_{i} C_i p^i, \tag{19}$$

Vectors p^i and coefficients C_i can be easily found numerically. Now, the vector v describing dynamics of the system after the jump can be written in the form

$$v = \sum_{i} C_i p^i e^{\lambda_i t}, \tag{20}$$

Where λ_i are the eigen-numbers of the matrix $G(d + \delta d)$. Dynamics of the absorbed power is given by the equation

$$P(t) = \omega(d + \delta d) \sum_{i=1}^{9} C_i e^{\lambda_i t} [p_5^i + p_6^i]$$
 (21)

The quantity $\delta P \equiv P(t) - P_0$ can be considered as a response of the system to the jump δd , and, at small δd , this response should be linear. The LIS we are interested in, is evidently proportional to the susceptibility $K(i\xi)$ connecting δP and δd . Then, if $S(\xi)$ is the Laplace transform of the response to the jump,

$$S(\xi) = \lim_{\alpha \to +0} \int_0^\infty e^{i\xi t} [P(t) - P_0] e^{-\alpha t} dt = -\omega (d + \delta d) \sum_{i=1}^9 \left[\frac{C_i(p_5^i + p_6^i)}{i\xi + \lambda_i} \right] + \frac{\omega d(b_5 + b_6)}{i\xi}, \tag{22}$$

then $|K(\imath\xi)|^2 = |\xi S(\xi)|^2$, and, for the LIS spectrum, we obtain

$$|\chi(\imath\xi)|^2 \sim |\xi S(\xi)|^2 \tag{23}$$

Thus, calculations of the LIS spectrum, within the simplest model described above can be performed using Eqs. (22) and (23).

IV. RESULTS

Let us calculate the LIS spectrum for essentially different relaxation rates in the optical transitions.

When the relaxation times entering the equations for elements of the density matrix (13) substantially exceed the period of optical nutation d, the dynamics of the density matrix for times smaller than the relaxation times is determined by Eq. (9). Under these conditions, this dynamics represents oscillations at frequencies equal to all possible differences between the eigen-values of the effective Hamiltonian W. The quantities W_i , i = 1, 2, 3, in this case, can be calculated either numerically or using Cardano's formula. Thus, the LIS spectrum will reveal peaks at appropriate frequencies, which, in this case, will be three: $\xi_1 \equiv |W_1 - W_2|, \xi_2 \equiv |W_1 - W_3|, \xi_3 \equiv |W_2 - W_3|$. This is demonstrated in Fig. 2(a), which shows the LIS spectrum obtained using Eqs. (22) and (23) for $d^{-1} << T_1, \tau_1, \tau_2$. Verical lines indicate frequencies ξ_i , i = 1, 2, 3.

In the absence of the magnetic field, one can easily see that $W_{1,2} = [\Delta \pm \sqrt{\Delta^2 + 4d^2}]/2$, $W_3 = 0$. At $d/\Delta << 1$, the LIS spectrum shows peaks at frequencies $\xi \approx \Delta$ and $\xi \approx d^2/\Delta$ (the peak is weakly pronounced), while at $d/\Delta << 1$, at the frequency of optical nutation 2d and at d (the peak is also weak).

The situation strongly changes when passing to short relaxation times of the optical transitions $\tau_2, \tau_1 \ll d^{-1}$. This case corresponds to the experiment [15] when the system becomes relatively transparent for the circularly polarized light with its intensity oscillating at the frequency of magnetic splitting (ESR frequency). In this case, the LIS spectrum shows a dip at the frequency of magnetic splitting 2ν [Fig. 2(b)]. The condition $\tau_2, \tau_1 \ll d^{-1}$ allows us to neglect the time derivatives $\dot{\sigma}_{13}, \dot{\sigma}_{31}, \dot{\sigma}_{23}, \dot{\sigma}_{32}, \dot{\sigma}_{33}$ in (13) as compared with the relaxation terms. This, in turn, makes it possible to express algebraically the matrix elements $\sigma_{13}, \sigma_{31}, \sigma_{23}, \sigma_{32}, \sigma_{33}$ pertinent to optical transitions through the elements $\sigma_{12}, \sigma_{21}, \sigma_{11}, \sigma_{22}$, related to the magnetically split doublet and, thus, to obtain the relationships similar to those presented in [15].

V. CONCLUSION

The goal of this paper is to show, using as an example a four-level system, potential usefulness and efficiency of

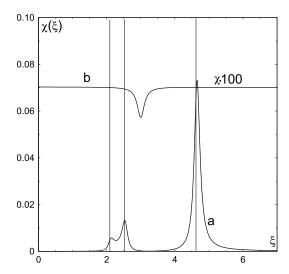


Figure 2. (a) the LIS spectrum for relatively long relaxation times in the optical transitions $\tau_2=10, \tau_1=2\tau_2$, (b) the same for relatively short relaxation times $\tau_2=0.01, \tau_1=2\tau_2$. The rest parameters are the same for both figures and are: $T_2=10, T_1=2T_2, d=1, \nu=\Delta=1.5$.

the "active" noise spectroscopy that employs transformation of intensity noise spectrum of the light passing through the medium as a source of information about its eigen-frequencies. This type of the noise spectroscopy has much in common with the conventional Faraday-rotation-based spin noise spectroscopy and, under certain conditions, provides the same information about the system and also may serve as a method of magnetic spectroscopy. At the same time, the technique considered

here, though deprived of the advantage of being nonperturbative, has a number of specific properties and unquestionable merits. In contrast to the Faraday-rotationbased technique, this method allows one to get information about optical transitions and properties of the excited states, rather than only about the ground-state dynamics and structure. The signal of the "active" noise spectroscopy, being a result of induced (rather than spontaneous) response of the system, can be controlled by the intensity of the optical field and, evidently, can be easier detected. This spectroscopy, unlike the "passive" FRbased, implies pure intensity-related measurements and, in the proposed version, does not require any polarimetric sensitivity. This technique employs fluctuations of the optical field rather than thermodynamic fluctuations of the ensemble of particles, and, therefore, the detected signal is not connected explicitly with the size of the light spot. It may seem strange, but a serious problem for this kind of spectroscopy may be the problem of laser source with a sufficiently high level of intensity noise in a sufficiently broad frequency range. Perhaps, special efforts will be needed to create a special laser source for the spectrometer of this kind. In principle, such a spectroscopy of the light intensity susceptibility can be implemented with harmonically (rather than randomly) modulated light. But in this case, to obtain the spectrum, we will have to scan the modulation frequency, and the experimental technique will lose its main advantage.

We consider the spectroscopy of the light intensity susceptibility to be a promising experimental technique for studying energy structure and dynamics of multilevel quantum systems under resonant optical excitation.

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