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Intensity-related optical susceptibility and ‘active’ noise spectroscopy

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Abstract

In this paper, we consider informative potentialities of the ‘active’ optical noise spectroscopy, under which we understand, generally, the spectroscopy of the response of a multilevel quantum system to a resonant optical field with its intensity (or polarization) modulated by a broadband (‘white’) noise. We show that, in the linear approximation, such a response can be treated most conveniently by introducing the notion of *Stokes susceptibility* (SS) whose spectrum is determined by a Laplace transform of the response to a small step-wise change of the light beam Stokes vector. The results of the calculations performed for a specific four-level energy diagram, typical for certain low-dimensional quantum systems, show that the low-frequency SS spectrum may provide information not only about the ground-state structure of the system (like conventional ‘passive’ spin noise spectroscopy), but also about the properties of the optical transitions (including nutation frequencies in the applied optical field). The considered version of the spin noise spectroscopy allows one, on the one hand, to take advantage of the highly efficient methods of data acquisition developed nowadays in this field of research and, on the other, to considerably broaden potentialities of the noise spectroscopy.

Keywords: noise spectroscopy, Rabi frequency, electron spin resonance

1. Introduction

The notion of susceptibility arises in a great variety of physical problems when describing the quantitative relationship between perturbation F of a material system and the response R it produces. The behavior of this quantity versus frequency forms the basis of most spectroscopic methods of research. The simplest (and, at the same time, the most important) is the case of *linear susceptibility* corresponding to the linear response of the system to perturbation with a harmonic time dependence. In this case, $F = F_\omega e^{-i\omega t}$ and $R = R_\omega e^{-i\omega t}$, with the susceptibility χ_ω being a complex proportionality factor connecting the amplitudes of the perturbation and response: $R_\omega = \chi_\omega F_\omega$. The spectroscopy of linear susceptibility provides information about the energy structure and relaxation characteristics of the system under study. An example of the linear susceptibility is the susceptibility ‘electric field—dipole moment’, with the electric field and dipole moment being the perturbation and response, respectively. The spectrum of this susceptibility at optical frequencies is being studied in the conventional linear optical spectroscopy. In this paper, we

describe the simplest properties of the susceptibilities ‘intensity—absorption’ and ‘polarization—absorption’. Here, as the perturbation we consider the intensity I of a polarized quasi-monochromatic optical wave with the frequency ω acting upon the system (atomic, molecular, or solid-state), and as the response, the energy P absorbed by the system per unit time. This susceptibility describes, in fact, the response of the system to small variations of the Stokes vector of the acting light (assuming its magnitude is proportional to the light intensity). This is the reason why we found it appropriate to use the term *Stokes susceptibility* (SS) to denote this quantity. Of course, the fact that the medium responds to the intensity-related characteristics of the light signifies its optical non-linearity. In this sense, the SS introduced here describes *linear* (or first-order) *nonlinearity* of the system.

It makes sense to note that the proposed SS spectroscopy can be treated, in a certain sense, as a spectroscopy of double resonance [1] because variations of the Stokes vector of the nominally monochromatic optical wave with frequency ω are unavoidably accompanied by the appearance of spectral sidebands with shifted frequencies, and the system proves to be

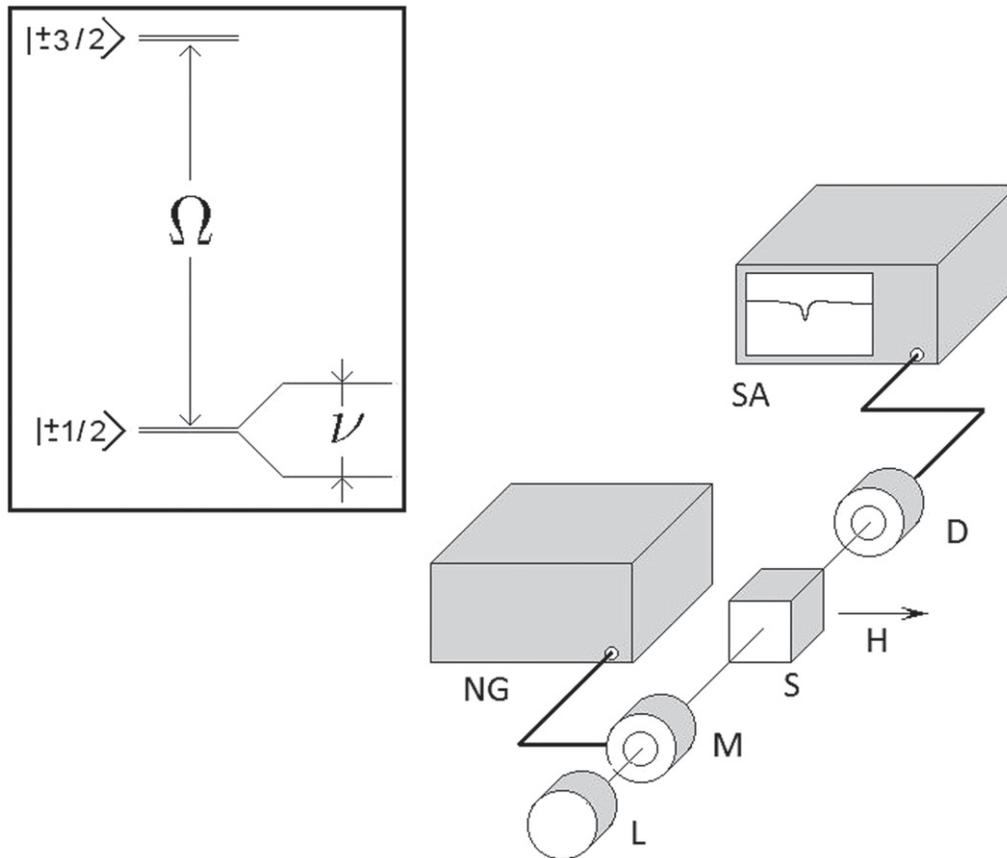


Figure 1. Schematic of experimental setup for observation of Stokes susceptibility (SS). L—laser, M—optical modulator, NG—noise generator, S—sample, H —transverse magnetic field, D—photodetector, SA—spectrum analyzer. Inset—energy structure of the model system.

probed by two optical fields, as it occurs in the double resonance spectroscopy. There are, however, essential distinctions between these two methods. First of all, the spectra of the double resonance, as a rule, reveals singularities in the vicinity of eigen-frequencies of the unperturbed Hamiltonian of the system, while the SS spectrum may reveal singularities at eigen-frequencies of some effective Hamiltonian that depends both on the energy structure of the system under study and on intensity and polarization of the quasi-monochromatic optical wave, used for SS detection.

Another specific feature of the SS susceptibility is of a methodological nature. The point is that the SS spectroscopy, like the spectroscopy of spin noise, implies the detection of signals in the range of radio-frequencies (up to a few GHz). This fact allows one to employ for the measurements the highly efficient and sensitive experimental equipment due to which the spin noise spectroscopy acquired in the last decade has shown high popularity [2, 5, 7–9].

A possible experimental setup for the observation of the SS spectrum with the use of noise spectroscopy equipment is shown in figure 1. The laser beam subjected to a broadband noise intensity/polarization modulation passes through the sample under study and hits a photodetector, whose output signal is fed to the input of the spectrum analyzer. If the intensity/polarization modulation spectrum of the laser beam is ‘white’, then we will observe, at the output of the spec-

trum analyzer, the SS spectrum of the studied sample. So, contrary to the conventional noise spectroscopy, which implies an observation of the spontaneous noise of the system, the SS spectroscopy implies an observation of the stimulated noise of the system and thus can be called ‘active’ noise spectroscopy.

The task of the present work can be formulated as follows. Assume that the intensity I of the quasimonochromatic electromagnetic field acting upon the system contains a dc (I_0) and a small ac ($\delta I(t)$) component $I = I_0 + \delta I(t)$, with the dc and ac components, having, in general, different polarizations. Correspondingly, the power absorbed by the system will contain the dc and ac components: $P = P_0 + \delta P(t)$. Then, the SS $K(i\nu)$ can be defined as $\delta P = K(i\nu)\delta I$ for the case $\delta I(t) \rightarrow \delta I e^{-i\nu t}$ and $\delta P(t) \rightarrow \delta P e^{-i\nu t}$. This is the quantity that is to be calculated for a specified model system. Below we will perform such calculations for a model system frequently encountered in semiconductor physics and will consider spectral properties of the SS in the range of relatively low (e.g., of the EPR range) frequencies $\nu \ll \omega$ (here ω is the frequency of the optical field). We will show that the SS of this model system contains information not only about transition frequencies and relaxation rates observed in linear spectroscopy, but also about frequencies of optical nutation (Rabi frequencies) usually obtained by means of pulsed pump-probe spectroscopy. We will see that, in contrast to the

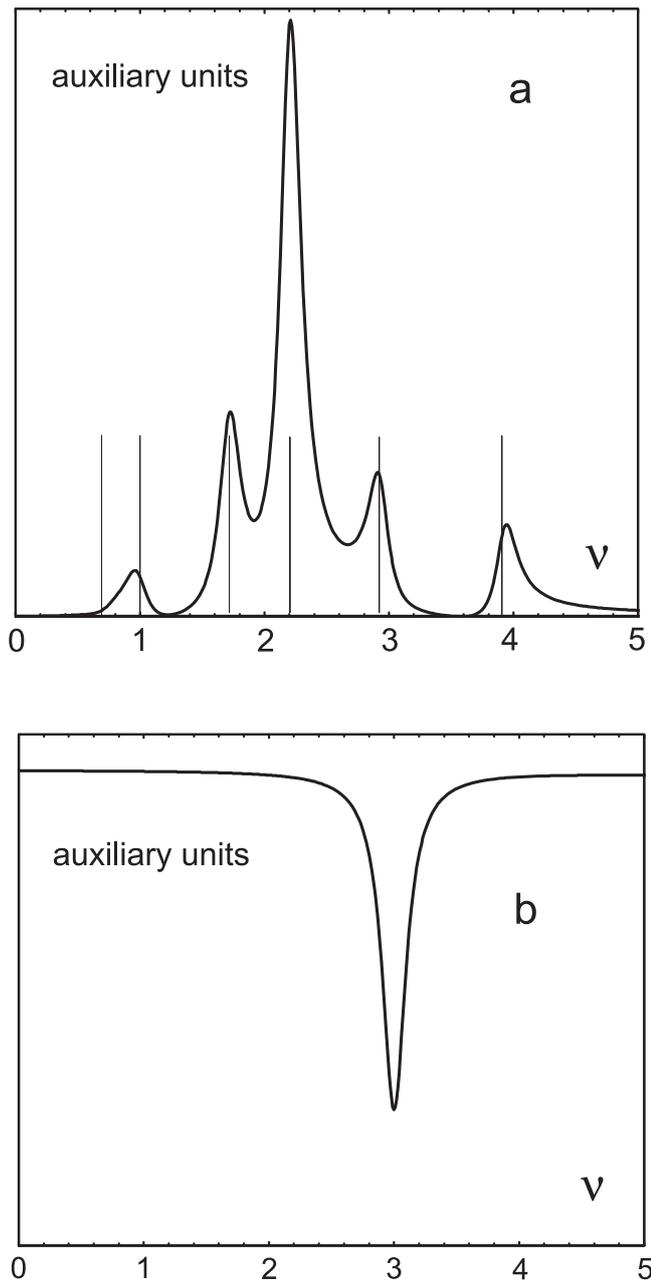


Figure 2. (a) frequency dependence of the intensity-related Stokes susceptibility for relatively long relaxation times in optical transitions: $\tau_2 = 10$, $\tau_1 = 2\tau_2$, the same for relatively short relaxation times: $\tau_2 = 0.01$, $\tau_1 = 2\tau_2$. The rest parameters, common for both plots, are: $T_2 = 10$, $T_1 = 2T_2$, $d = 1$, $\nu = \Delta = 1.5$.

conventional optical spectroscopy, which allows one to measure frequencies of transitions between the states of the nonperturbed Hamiltonian of the system, the SS spectroscopy makes it possible (in a certain range of relaxation rates) to measure the frequencies of transitions between the states of the effective intensity-dependent Hamiltonian¹.

¹ In the case of a two-level system, this is the total Hamiltonian of the problem that takes into account the time-dependent electromagnetic field written in the coordinate frame rotating with optical frequency ω .

In spite of the fact that the ‘active’ noise spectroscopy, which implies real optical excitation of the system with real dissipation of the absorbed energy, is essentially *perturbative*, it has very much in common with the spectroscopy of spin noise (see, e.g., [2]), which uses a highly stable probe beam, propagating through a transparent sample, detects *spontaneous* fluctuations of its birefringence caused by its magnetization noise, and is considered, to a great extent, nonperturbative. In principle, such a nonperturbative approach can be also realized with the probe beam propagating in the region of optical absorption [3]. In the SS spectroscopy, on the contrary, the response is fundamentally induced by modulation of the light beam and cannot be observed for the light with a fixed Stokes vector. The first observation of resonant response in the SS spectrum was, in fact, described by Bell and Bloom in 1961 [4], long before the first experiment on spin noise spectroscopy [5]. The authors of [4] used, in their study, the light beam with a monochromatically (rather than randomly) modulated Stokes vector. The experiments of this kind with the noise-modulated light were described more recently in [3], where the resonant paramagnetic susceptibility of an atomic gas was detected in this way. The response of a two-level and three-level system to the noise-modulated optical excitation was studied theoretically in [3, 6, 10–13] where it has been shown, in particular, that the SS spectrum may reveal spectral features related to Rabi frequencies of the system.

It should be noted that the scheme with monochromatically modulated light employed by Bell and Bloom [4] is ideologically identical to the noise-assisted SS spectroscopy, but from the technical viewpoint they differ drastically due to possibility of the up-to-date electronics to detect a response on all available frequencies at a time (rather than on a single one). The recent advent of the ultrafast digital spectrum analyzers with the operation bandwidth of ~ 1 GHz and the averaging of the Fourier spectrum in real time made it possible to greatly increase the sensitivity of the noise spectroscopy. This technical advancement has played a crucial role in the development of the present-day spin noise spectroscopy [7, 8] and can be evidently applied to the ‘active’ SS noise spectroscopy described in our paper.

It is noteworthy that the experimental approach analyzed in this paper refers to the broad field of research associated with spectroscopy of driven atoms intensely developed in the 1970s–1980s (see, e.g., [14–16]). An essential distinctive feature of the technique considered here is that it implies monitoring the system by detecting distortion of the light *intensity spectrum*, rather than distortion of the optical spectrum of the system with the aid of a separate probe beam. This is why the ‘active’ spectroscopy of optical noise, when the pump laser beam modulated by a broadband noise serves simultaneously as a probe, occupies a special niche in the nonlinear optical spectroscopy [3, 6, 17–19].

It is also important that the experiment under consideration can be performed with pure polarization modulation of the probe light (keeping the light intensity constant). In this case, the intensity noise of the light after the sample is related only to its nonzero SS and is detected with no

background (unlike the case of noise intensity modulation). For this reason, this version of the experiment may provide higher sensitivity in the SS measurements.

In the next Section, we calculate the SS of a simplest model system and analyze its frequency dependence.

2. Model calculations

2.1. Starting points

The sample under study is assumed to represent an ensemble of particles, so that the power absorbed by the sample irradiated by the electromagnetic field is equal to the sum of powers absorbed by each particle separately. The energy spectrum of an individual particle (figure 1, inset) is supposed to be comprised of two doublets, namely, the ground one $\pm 1/2$ and the excited one $\pm 3/2$ (the indicated quantities are components of angular momentum upon the direction coincident with the z -axis of our coordinate system). Magnetic splitting of the ground doublet $\pm 1/2$ is characterized by an isotropic g -factor, while the doublet $\pm 3/2$ can be split only by the field component parallel to the z -axis. The spectral distance between the two doublets (denoted by Ω) is assumed to be close to the frequency of the acting optical field ω . We suppose that Ω lies in the optical range and substantially exceeds all other frequency (energy) parameters of the problem.

It should be noted that this energy diagram, with its specific characteristics, is typical for certain two-dimensional semiconductor structures [20] and is therefore of great interest for the present-day semiconductor optics. The widely used model of a charged quantum dot can serve as an example of the above four-level system. In this case, the ground doublet corresponds to two possible spin orientations of the resident electron, while the excited one corresponds to the same for the hole.

The SS introduced above was calculated in the following way. Initially, the system under study was supposed to be irradiated, for a sufficiently long time, by a monochromatic light with a fixed intensity and polarization, so that the power absorbed by the system is stationary. Then, the Stokes vector of the light (its intensity or polarization) experiences a small jump. The response of the system to this jump was used to find the sought susceptibility by means of the Laplace transform.

2.2. The dynamics of the system with no relaxation

To fulfil the program specified in the previous subsection, we will reproduce, to a considerable extent, the conditions of the experiment [4] and will consider the system in a transverse (directed along the x -axis) magnetic field with the strength \mathcal{H} (figure 1). The optical field will be considered as a sum of a linearly polarized wave with the amplitude A and a circularly polarized wave with the amplitude B propagating along the z -

axis. The relevant Hamiltonian has the form:

$$H_1 = A \cos \omega t [S_x \cos \alpha + S_y \sin \alpha] + B [S_x \cos \omega t + S_y \sin \omega t] \quad (1)$$

Here, α is the angle specifying the direction of the linearly polarized component of the optical field and $S_{x,y}$ are the operators that have the same nonzero matrix elements as the operators of the appropriate components of the angular momentum. Let us arrange the states of the model system in the following order $|+1/2\rangle | -1/2\rangle | -3/2\rangle | +3/2\rangle$. Then, the matrices H_1 and nonperturbed Hamiltonian H_0 will acquire the form

$$H_1 = \frac{A \cos \omega t}{2} \begin{pmatrix} 0 & 0 & 0 & \xi \\ 0 & 0 & \bar{\xi} & 0 \\ 0 & \xi & 0 & 0 \\ \bar{\xi} & 0 & 0 & 0 \end{pmatrix} + \frac{B}{2} \begin{pmatrix} 0 & 0 & 0 & \xi_1 \\ 0 & 0 & \bar{\xi}_1 & 0 \\ 0 & \xi_1 & 0 & 0 \\ \bar{\xi}_1 & 0 & 0 & 0 \end{pmatrix} \quad (2)$$

$$\xi \equiv e^{i\alpha} \quad \xi_1 \equiv e^{i\omega t}$$

$$H_0 = \begin{pmatrix} 0 & \nu & 0 & 0 \\ \nu & 0 & 0 & 0 \\ 0 & 0 & \Omega & 0 \\ 0 & 0 & 0 & \Omega \end{pmatrix} \quad \nu \equiv \frac{g\beta\mathcal{H}}{2\hbar} \quad (3)$$

It is convenient first to pass to the basis where H_0 is diagonal and then, to the rotating coordinate frame, where the total Hamiltonian is time-independent. The first step corresponds to the unitary transformation with the following matrix

$$S \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & -1 & 0 & 0 \\ 0 & 0 & \sqrt{2} & 0 \\ 0 & 0 & 0 & \sqrt{2} \end{pmatrix}, \quad \text{with } S^{-1} = S \quad (4)$$

Let us supply by tilda all the operators of the form $\tilde{H} \equiv SHS$. Then, as can be shown by direct calculations,

$$\tilde{H}_0 = \begin{pmatrix} \nu & 0 & 0 & 0 \\ 0 & -\nu & 0 & 0 \\ 0 & 0 & \Omega & 0 \\ 0 & 0 & 0 & \Omega \end{pmatrix} \quad (5)$$

$$\tilde{H}_1 = \frac{A \cos \omega t}{2\sqrt{2}} \begin{pmatrix} 0 & 0 & \bar{\xi} & \xi \\ 0 & 0 & -\bar{\xi} & \xi \\ \xi & -\bar{\xi} & 0 & 0 \\ \bar{\xi} & \bar{\xi} & 0 & 0 \end{pmatrix} + \frac{B}{2\sqrt{2}} \begin{pmatrix} 0 & 0 & \bar{\xi}_1 & \xi_1 \\ 0 & 0 & -\bar{\xi}_1 & \xi_1 \\ \xi_1 & -\bar{\xi}_1 & 0 & 0 \\ \bar{\xi}_1 & \bar{\xi}_1 & 0 & 0 \end{pmatrix} \quad (6)$$

and the density matrix $\tilde{\rho} = S\rho S$ describing the state of the system satisfies the equation $i\partial\tilde{\rho}/\partial t = [\tilde{H}, \tilde{\rho}]$. Now, we pass to the rotating frame using the following time-dependent matrix transformation:

$$\hat{H} = e^{iMt} \tilde{H} e^{-iMt} \quad \sigma \equiv e^{iMt} \tilde{\rho} e^{-iMt} \quad (7)$$

where the diagonal matrix M has the form

$$M \equiv \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & \omega & 0 \\ 0 & 0 & 0 & \omega \end{pmatrix} \quad (8)$$

Then, one can easily find that the matrix σ meets the equation $i\dot{\sigma} = [\hat{H} - M, \sigma]$. Calculation of the matrix \hat{H} shows that it contains time-independent elements as well as those proportional to $e^{\pm 2i\omega t}$. The matrix elements oscillating at double frequency are ignored as essentially nonresonant. Taking into account these remarks, the matrix \hat{H} and equation for the density matrix acquire the form:

$$\hat{H} = \begin{pmatrix} \nu & 0 & \bar{\xi}d & \xi d + d_1 \\ 0 & -\nu & -\bar{\xi}d & \xi d + d_1 \\ \xi d & -\xi d & \Omega & 0 \\ \bar{\xi}d + d_1 & \bar{\xi}d + d_1 & 0 & \Omega \end{pmatrix} \quad (9)$$

where

$$d \equiv \frac{A}{4\sqrt{2}} \quad d_1 \equiv \frac{B}{2\sqrt{2}}$$

Now all the operators are presented by the matrices 4×4 , and the equation of motion for the density matrix σ has the form:

$$i\dot{\sigma} = [W, \sigma], \quad W \equiv \hat{H} - M = \begin{pmatrix} \nu & 0 & \bar{\chi} & \chi_1 \\ 0 & -\nu & -\bar{\chi} & \chi_1 \\ \chi & -\chi & \Delta & 0 \\ \bar{\chi}_1 & \bar{\chi}_1 & 0 & \Delta \end{pmatrix} \quad (10)$$

where

$$\Delta \equiv \Omega - \omega\chi \equiv de^{i\alpha} \quad \chi_1 \equiv de^{i\alpha} + d_1$$

Our task is to calculate the power absorbed by the system. This power can be written as follows:

$$P = \frac{d}{dt} \text{Sp } \rho H = \frac{d}{dt} \text{Sp } \sigma \hat{H} = \text{Sp } \dot{\sigma} \hat{H} + \text{Sp } \sigma \frac{d\hat{H}}{dt} \quad (11)$$

Using the equations for the matrices \hat{H} and M , we eventually have:

$$P = 2\omega d \text{Im} [\bar{\xi}(\sigma_{32} - \sigma_{31} + \sigma_{14} + \sigma_{24})] + 2\omega d_1 \text{Im} [\sigma_{14} + \sigma_{24}] \quad (12)$$

Equation (10) connects the density matrix elements σ_{ik} , $i, k = 1, 2, 3, 4$ and their time derivatives. Let us introduce the vector-column u_α , $\alpha = 1, 2, \dots, 16$, whose components are the elements of the density matrix numbered by rows: $u_\alpha = \sigma_{ik}$, $\alpha = 4(i-1) + k$. For instance, $\sigma_{11} = u_1$, $\sigma_{22} = u_6$, $\sigma_{21} = u_5$... For a given number $1 < \alpha < 16$, one can find the first i_1 and second i_2 indices: $i_1 = F_1(\alpha)$, $i_2 = F_2(\alpha)$, where the integer functions $F_{1,2}(\alpha)$ can be defined by the tables. For instance,

$F_1(1) = 1$, $F_1(7) = 3$, $F_2(3) = 1$. It is clear that the equation (10) for the density matrix is equivalent to an equation for the 16-component vector u :

$$\dot{u} = Gu \quad (13)$$

Where G is the 16×16 matrix. To express matrix G 16×16 in terms of matrix W 4×4 , let us define the function $W(i, k)$ of two variables i and k so that at integer i and k it coincides with the corresponding matrix element of the Hamiltonian W and equals zero otherwise. Then, the calculation gives the following expression for the matrix G :

$$G_{\alpha\beta} = i \left[W(\beta - 4[F_1(\alpha) - 1], F_2(\alpha)) - W\left(F_1(\alpha), \frac{\beta - F_2(\alpha)}{4} + 1\right) \right] \quad (14)$$

$\alpha, \beta = 1, 2, \dots, 16$

2.3. The relaxation and steady state regime

To obtain the steady-state regime under optical excitation, we have to take into account relaxation processes. The relaxation of non-diagonal elements of the density matrix (dephasing) can be taken into consideration by the replacement $G_{\alpha\alpha} \rightarrow G_{\alpha\alpha} - 1/\tau_\alpha$ with $\alpha \neq 1, 6, 11, 16$. Besides, since the relaxation times of the non-diagonal matrix elements σ_{ik} and σ_{ki} are the same, the following relation should be satisfied: $\tau_\alpha = \tau_{4[F_2(\alpha)-1]+F_1(\alpha)}$.

To consider the relaxation of diagonal elements of the density matrix, the equations for them should be modified as follows:

$$\begin{cases} \dot{\sigma}_{11} = \dots \frac{\sigma_{22} - \sigma_{11}}{T_1^l} + \frac{\sigma_{33}}{T_{13}} + \frac{\sigma_{44}}{T_{14}} \\ \dot{\sigma}_{22} = \dots \frac{\sigma_{11} - \sigma_{22}}{T_1^l} + \frac{\sigma_{33}}{T_{23}} + \frac{\sigma_{44}}{T_{24}} \\ \dot{\sigma}_{33} = \dots \frac{\sigma_{44} - \sigma_{33}}{T_1^u} - \frac{\sigma_{33}}{T_{13}} - \frac{\sigma_{33}}{T_{23}} \\ \dot{\sigma}_{44} = \dots \frac{\sigma_{33} - \sigma_{44}}{T_1^u} - \frac{\sigma_{44}}{T_{14}} - \frac{\sigma_{44}}{T_{24}} \end{cases}$$

or in terms of the u -vector

$$\begin{cases} \dot{u}_1 = \dots \frac{u_6 - u_1}{T_1^l} + \frac{u_{11}}{T_{13}} + \frac{u_{16}}{T_{14}} \\ \dot{u}_6 = \dots \frac{u_1 - u_6}{T_1^l} + \frac{u_{11}}{T_{23}} + \frac{u_{16}}{T_{24}} \\ \dot{u}_{11} = \dots \frac{u_{16}}{T_1^u} - u_{11} \left[\frac{1}{T_1^u} + \frac{1}{T_{13}} + \frac{1}{T_{23}} \right] \\ \dot{u}_{16} = \dots \frac{u_{11}}{T_1^u} - u_{16} \left[\frac{1}{T_1^u} + \frac{1}{T_{14}} + \frac{1}{T_{24}} \right] \end{cases} \quad (15)$$

$$\Delta G_{1,1} = -\frac{1}{T_1^l}, \quad \Delta G_{1,6} = \frac{1}{T_1^l},$$

$$\Delta G_{1,11} = \frac{1}{T_{13}}, \quad \Delta G_{1,16} = \frac{1}{T_{14}}$$

$$\Delta G_{6,1} = \frac{1}{T_1^l}, \quad \Delta G_{6,6} = -\frac{1}{T_1^l},$$

$$\Delta G_{6,11} = \frac{1}{T_{23}}, \quad \Delta G_{6,16} = \frac{1}{T_{24}}$$

$$\Delta G_{11,11} = -\frac{1}{T_1^u} - \frac{1}{T_{13}} - \frac{1}{T_{23}}, \quad \Delta G_{11,16} = \frac{1}{T_1^u}$$

$$\Delta G_{16,11} = \frac{1}{T_1^u}, \quad \Delta G_{16,16} = -\frac{1}{T_1^u} - \frac{1}{T_{14}} - \frac{1}{T_{24}}$$

The ellipsis here implies terms of the dynamic equations with no regard to the relaxation. The quantities T_1^u and T_1^l have the sense of the population relaxation times for the upper and lower doublets, respectively. T_{13} is the radiative decay time for the transitions 1–3. Similarly, we assume that $T_{13} = 2\tau_3$. In terms of the vector u , the equation for the absorbed power (12) takes the form

$$P = 2\omega d \operatorname{Im} \left[\bar{\xi} (u_{10} - u_9 + u_4 + u_8) \right] + 2\omega d_1 \operatorname{Im} [u_4 + u_8] \quad (16)$$

As it was mentioned above our task is to calculate the response of the system to a jump of the pump: $d \rightarrow d + \delta d$ and $d_1 \rightarrow d_1 + \delta d_1$ (remind that d and d_1 are the normalised amplitudes (9) of linearly and circularly polarised components of the optical field). To solve this problem, we have to calculate the steady-state density matrix before the jump at given values of d and d_1 . This matrix can be found from the equation $G(d, d_1)u = 0$. Thus, the matrix $G(d, d_1)$ should always have a zero eigen value. We will denote the eigen vector corresponding to this value by e^{16} . To obtain the vector $b \sim e^{16}$ that described the steady-state density matrix, the vector e^{16} should be normalized as $b_1 + b_6 + b_{11} + b_{16} = 1$ and consequently

$$b = \frac{e^{16}}{e_1^{16} + e_6^{16} + e_{11}^{16} + e_{16}^{16}}$$

In this case, in conformity with equation (16), the power absorbed before the jump is given by the formula

$$P_0 = 2\omega d \operatorname{Im} \left[\bar{\xi} (b_{10} - b_9 + b_4 + b_8) \right] + 2\omega d_1 \operatorname{Im} [b_4 + b_8] \quad (17)$$

2.4. The jump

The jump of the intensity and polarization of the optical field corresponds to $d \rightarrow d + \delta d$ and $d_1 \rightarrow d_1 + \delta d_1$. Let the vectors p^i and numbers $\lambda^i, i = 1, \dots, 16$ be eigen for the matrix $G(d + \delta d, d_1 + \delta d_1)$. To find the transient response, we have to solve the equation $\dot{u} = G(d + \delta d, d_1 + \delta d_1)u$ at the initial condition $u(t = 0) = b$. Let us decompose the vector over eigen vectors of the matrix

$$b = \sum_1^{16} C_i p^i \quad (18)$$

The coefficients C_i are found numerically. Then, solution of the problem acquires the form:

$$u(t) = \sum_{i=1}^{16} C_i p^i e^{\lambda_i t} \quad (19)$$

The absorbed power after the jump is given by the expression

$$P(t) = 2\omega(d + \delta d) \operatorname{Im} \left[\bar{\xi} \sum_{i=1}^{16} C_i (p_{10}^i - p_9^i + p_4^i + p_8^i) e^{\lambda_i t} \right] + 2\omega(d_1 + \delta d_1) \operatorname{Im} \left[\sum_{i=1}^{16} C_i (p_4^i + p_8^i) e^{\lambda_i t} \right] \quad (20)$$

Let us define the quantity S as

$$S(\nu) = \lim_{\alpha \rightarrow +0} \int_0^\infty e^{\nu t} [P(t) - P_0] e^{-\alpha t} dt \quad (21)$$

$$= i\omega(d + \delta d) \sum_{i=1}^{16} \left\{ \frac{Z_i}{i\nu + \lambda_i} - \frac{\bar{Z}_i}{i\nu + \bar{\lambda}_i} \right\} + \frac{2\omega d}{i\nu} \operatorname{Im} \left[\bar{\xi} (b_{10} - b_9 + b_4 + b_8) \right] + i\omega(d_1 + \delta d_1) \sum_{i=1}^{16} \left\{ \frac{Y_i}{i\nu + \lambda_i} - \frac{\bar{Y}_i}{i\nu + \bar{\lambda}_i} \right\} + \frac{2\omega d_1}{i\nu} \operatorname{Im} [b_4 + b_8]$$

where

$$Z_i \equiv \bar{\xi} C_i (p_{10}^i - p_9^i + p_4^i + p_8^i) \\ Y_i \equiv C_i (p_4^i + p_8^i)$$

Then, the module squared of the sought SS is given by the expression

$$|K(i\nu)|^2 = |\nu S(\nu)|^2 \quad (22)$$

3. Discussion

When the relaxation times entering the equations for elements of the density matrix $\dot{u} = Gu$ essentially exceed the optical nutation period $\chi^{-1}, \chi_1^{-1} \ll T_1^u, T_1^l, T_{13}, T_{14}, T_{23}, T_{24}, \tau_\alpha$, the dynamics of the density matrix for times shorter than the relaxation times is determined by equation (10). Under these conditions, the above dynamics represent oscillations at frequencies corresponding to all possible differences of the appropriate eigen values $W_i, i = 1, 2, 3, 4$ of the effective Hamiltonian W (equation (10)), with the values of $W_i, i = 1, 2, 3, 4$ calculated numerically. Therefore, the frequency dependence of the SS will reveal peaks at appropriate frequencies, which, in this case, will be six: $\xi_1 \equiv |W_1 - W_2|, \xi_2 \equiv |W_1 - W_3|, \xi_3 \equiv |W_1 - W_4|, \xi_4 \equiv |W_2 - W_3|, \xi_5 \equiv |W_2 - W_4|, \xi_6 \equiv |W_3 - W_4|$. This is demonstrated in figure 2(a), which shows frequency dependences of the SS obtained using equations (21) and (22) at $d = 0.4, \delta d = 0.1d, d_1 = 0.3, \delta d_1 = 0.1d_1, \Delta = 1, \nu = 1.5$. All the dephasing times and all the population relaxation times were taken to be 10 and 20, respectively. Vertical lines show frequencies $\xi_i, i = 1, \dots, 6$ obtained by numerical diagonalization of matrix (10).

The situation essentially changes when we pass to shorter relaxation dephasing times $\tau_\alpha \ll \chi_1^{-1}, \chi^{-1}$, (with α

corresponding to the optical transitions $1 \rightarrow 3$, $1 \rightarrow 4$, $2 \rightarrow 3$, $2 \rightarrow 4$). This case corresponds to the experiment of Bell and Bloom [5], when the system becomes transparent to the light whose intensity oscillates at the frequency of magnetic splitting 2ν (EPR frequency). In this case, the SS should reveal a dip at the frequency of magnetic splitting 2ν . This is illustrated by figure 2(b), which shows the frequency dependence of the SS calculated for the case when the above relaxation times of the optical transitions were taken by a factor of 1000 shorter than in figure 2(a). It should be emphasized that the dephasing time of the transition $1 \rightarrow 2$, which controls the width of the SS resonance at the frequency 2ν , remained, as before, equal to 10.

Finally, consider the possibility, mentioned in the introduction, to detect the SS spectrum using the light beam with a fixed intensity. In this case, the quantities δd and δd_1 should be connected by the following condition

$$\delta d_1 = -\delta d \frac{2d + d_1 \cos \alpha}{d_1 + d \cos \alpha} \quad (23)$$

(see definition (9) for the normalised amplitudes d and d_1 of the linearly and circularly polarised waves). Under this condition, the SS spectrum appears to be similar to that described above. For instance, in the case of short dephasing times in the optical transitions (as in the experiment of Bell and Bloom [5]), with the satisfied condition (23) (when only *polarization* of the incident light is modulated), frequency dependence of the SS appears to be qualitatively similar to that shown in figure 2(b).

4. Conclusions

In this paper we analyze informative potentialities of the ‘active’ noise spectroscopy that implies studying the frequency dependence of the response of a system to small variations of the light beam Stokes vector. We show that this *Stokes spectroscopy*, unlike the Faraday-rotation-based spectroscopy of spontaneous spin noise, does not require such high polarimetric sensitivity and, therefore allows one to obtain easier signals of resonant response with uncontrollable magnitude. The drawback of this spectroscopy of being essentially perturbative is compensated by additional possibilities to detect frequencies of the light-induced dynamics of the system usually derived from transients of nonlinear

optical response. The noise-assisted spectroscopy of Stokes susceptibility looks especially promising in the context of the up-to-date high-speed methods of data acquisition and processing.

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