Nonlinear photoprocesses in bichromophores. II. Correlated population and fluorescence intensity fluctuations

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We consider sationary fluctuations due to nonlinear excitation in a bichromophore, that is, in a molecule consisting of two chromophoric groups. We describe correlated fluctuations of the level populations and of the flux of flourescent photons emitted by a bichromophore. The population fluctuations are observable in experiments involving quasielastic particle scattering or Raman scattering of probe radiation. Experiments of this kind can yield information about the rate of nonradiative energy transfer. The photon correlation is characterized by antibunching and sub-Poissonian statistics and can yield information about collective excitation and nonlinear quenching.

(Abstract translated from Russian by V.A.Markel)

Nonlinear photoprocesses in bichromophores. II. Correlated population and fluorescence intensity fluctuations

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Stationary fluctuations in a bichromophore (a molecule containing two chromophore groups) accompanying its nonlinear interaction are examined. The correlation fluctuations in the level populations of the two monomers comprising the bichromophore are described together with the flux of fluorescence photons emitted by the bichromophore. The population fluctuations are measured in experiments on quasielastic particle scattering as well as Raman scattering of probe radiation. These experiments make it possible to obtain information on the nonradiative energy exchange rates between monomers. The photon correlation has an antigrouping character and is responsible for the photon sub-Poisson statistics. It contains information on the cooperative excitation (nonlinear quenching) in the bichromophore.

INTRODUCTION

Reference 1 examines the nonlinear optical excitation of a bichromophore: a molecule containing two chromophore groups. Kinetic (balance) equations are obtained and are used to find the average populations.

The present study is devoted to a theoretical description of stationary (i.e., with fixed excitation conditions) fluctuations in an ensemble of bichromophore molecules. These fluctuations are caused by the randomness of the interlevel transitions (the kinetic equations describe only the transition probabilities and do not determine them). Excitation exchange causes the population fluctuations in each of the monomers in the bichromophore to be correlated. The autocorrelation function of fluorescence intensity fluctuations is determined. Sub-Poisson photon statistics and an antigrouping effect are predicted.

CORRELATED STATIONARY POPULATION FLUCTUATIONS

Since n_{11} is the probability that both monomers of a bichromophore are located in the lower excited states S_1, it is possible to find the (simultaneous) correlation factor of the populations of these states from stationary solutions of kinetic equations (1.25), (1.29)[1)]

$$K\equiv\frac{n_{11}-n_1^2}{n_1^2}=-\frac{\beta_{11}}{2}\frac{\Gamma_1+\frac{\beta_{11}}{2}+[r_0(\sigma_{21}-\sigma_{10})+\sigma_{01}]I}{\left[\Gamma_1+\frac{\beta_{11}}{2}+(\sigma_{10}+\bar\sigma_1)I\right]^2} \tag{1}$$

It is clear therefore that when $I<[\Gamma_1+(\beta_{11}/2)]/[r_0(\sigma_{10}-\sigma_{21})-\sigma_{01}]$ we have population anticorrelation, while in the opposite case there is a positive correlation. If $r_0(\sigma_{10}-\sigma_{21})-\sigma_{01}\leqslant 0, K<0$ for all I. The population correlation and excitation energy transport between monomers have a self-consistent determinate relationship.[1]

In order to describe the fluctuation dynamics we will introduce the microscopic populations—the functions $v_{ij}(t)$—in the following manner: $v_{ij}(t)=1$ if at time t the first monomer in the given bichromophore is in state $S_i^{\rm I}$, while the second is in $S_j^{\rm II}$, and in the opposite case $v_{ij}(t)=0$. The population n_{ij} and the correlation function of the populations of the bichromophore $P_{ij}^{kl}(\tau)$ are expressed as the average over the ensemble

$$n_{ij}=\langle\nu_{ij}(t)\rangle,\quad P_{ij}^{kl}(\tau)=\langle\nu_{ij}(t)\nu_{kl}(t+\tau)\rangle=\langle\nu_{ij}(0)\nu_{kl}(\tau)\rangle. \tag{2}$$

The function $P_{ij}^{kl}(\tau)$ in Eqs. (2) is the joint probability that the bichromophore at time $t=0$ is in state (i,j) and $t=\tau$ in state (k,l). When $\tau>0$, $P_{ij}^{kl}(\tau)$ satisfies kinetic equation system (1.18)–(1.20) with respect to the upper pair of indices, while the lower pair of indices represent the initial conditions: $P_{ij}^{kl}(0)=\delta_{ik}\delta_{jl}n_{ij}$. It follows from these properties that $P_{ij}^{kl}(\tau)$ is proportional to the Green's function G_{ij}^{kl} of system (1.18)–(1.20)

$$P_{ij}^{kl}(\tau)=n_{ij}G_{ij}^{kl}(\tau),\quad G_{ij}^{kl}(0)=\delta_{ik}\delta_{jl}. \tag{3}$$

Equations (3) are an expression of the Onsager principle. These solve the problem of describing the dynamics of the correlated fluctuations in principle.

We will introduce the microscopic level populations $\nu_1^{\rm I}\nu_1^{\rm II}$ of the separate chromophores $\nu_1^{\rm I}=\sum\limits_{i=0}^{2}\nu_{1i},\nu_1^{\rm II}=\sum\limits_{i=0}^{2}\nu_{i1}$ through which the mutual correlation function of the level population fluctuations of the monomers $C(\tau)$ and the analogous autocorrelation function $\tilde C(\tau)$ are expressed

$$\begin{aligned}C(\tau)&=\langle\nu_1^{\rm I}(0)\nu_1^{\rm II}(\tau)\rangle-n_1^2=\sum_{i,\,j=0}^{1}P_{j1}^{1i}(\tau)-n_1^2,\\ \tilde C(\tau)&=\langle\nu_1^{\rm I}(0)\nu_1^{\rm I}(\tau)\rangle-n_1^2=\sum_{i,\,j=0}^{1}P_{j1}^{i1}(\tau)-n_1^2.\end{aligned} \tag{4}$$

The minor contributions associated with the population of the S_2 levels are dropped from Eq. (4).

The expression for the Green's functions in Eqs. (3) is very cumbersome. Considering a symmetric bichromophore for simplicity in the limiting case of strong cooperative processes $(\beta_{11}\gg\Gamma_1,\sigma_{10},I)$ and the rapid decay of the level $S_2(\beta_{20},\beta_{21}\ll\Gamma_2)$ we obtain

$$\begin{aligned}C(\tau),\ \tilde C(\tau)&=\frac{n_1}{2}\exp\{-[\Gamma_1+(\sigma_{10}+\bar\sigma_1)I]\tau\}\\ &\times[\mp\exp(-2\beta_{01}\tau)+(1-2n_1)\exp(-(1+r_0)\sigma_{10}I\tau)],\end{aligned} \tag{5}$$

where the plus applies to $\tilde C(\tau)$, and the minus applies to $C(\tau)$; n_1 is given by Eq. (1.29).

Figure 1 shows the behavior of the mutual correlation function $C(\tau)$ [Eq. (5)] for various excitation exchange rates $\beta_{10}=\beta_{01}$ [Eq. (1.5)]. With small delays τ there will always be an anticorrelation of the populations [compare to Eq. (1)] which can be attributed to cooperative quenching.

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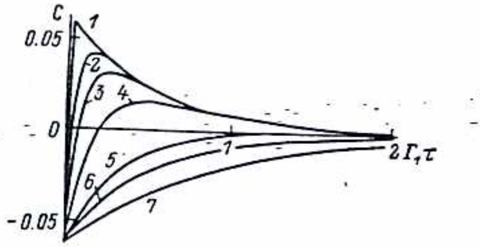

FIG. 1. Irreducible correlation function $C(\tau)$ [Eq. (5)] of the first level populations. Plotted for a symmetrical bichromophore with a relative intensity $I/I_1 = 0.5$; $r_0 = 0$, $\bar{\sigma}_1 = 0$ in the limit $\beta_{11} \gg \Gamma_1$. The relative exchange transfer rates $\beta_{10}/\Gamma_1 = 100, 10, 5, 2, 0.5, 0.25, 0$ (curves 1–7).

The latter (under these conditions) will cause one of the two monomers on the S_1 levels to be excited to the S_2 state from which it immediately returns to S_1, while the other jumps to S_0. Thus the level populations of the various monomers fluctuate in antiphase. The reversal of the correlation sign with increasing τ can be attributed to the fact that the population process from one monomer due to exchange processes (1.5) is transferred to the other monomer. Unlike cross-correlation function $C(\tau)$ autocorrelation function $\widetilde{C}(\tau)$ [Eq. (5)] is positive, and monotonically diminishes in τ.

FLUORESCENCE INTENSITY FLUCTUATIONS AND SUB-POISSON PHOTON STATISTICS

For definiteness we will consider the integrated fluorescence over the monomers. We will assume that the spectral fluorescence width $\Delta\omega$ is sufficiently great (much greater than $\Gamma_1, \sigma_{10}I$). We can therefore neglect the coherent (Gaussian) fluctuations in the number of photons, whose correlation time is $1/\Delta\omega$ with the times characteristic of this study. Gaussian fluctuations are also eliminated if the photodetector area significantly exceeds the coherence area. We will consider non-Gaussian fluctuations (for their properties see, for example, Ref. 2) associated with the finite number of radiators.

Let $J(t)$ be the photon flux (the number of quanta incident on the photodetector per unit time). The normalized autocorrelation function $g(\tau)$ of the photon flux under conditions of non-Gaussian fluctuations is expressed through such a function for $J_{\bar{1}}(t)$: The photon flux at the photodetector emitted by a single particle (by the bichormophore)

$$g(\tau) \equiv \frac{\langle J(0)J(\tau)\rangle - \langle J\rangle^2}{\langle J\rangle^2} = \frac{f(\tau)}{N},$$

$$f(\tau) = \frac{\langle J_1(0)J_1(\tau)\rangle - \langle J_1\rangle^2}{\langle J_1\rangle^2}, \tag{6}$$

where N is the number of bichromophores from which radiation is recorded. The average flux is expressed as $\langle J_1\rangle = 2\delta Q\Gamma_1 n_1$, where Q is the flouoresence quantum efficiency; δ is a geometric factor (the fraction of the total number of photons striking the photodetector).

The autocorrelation function of the photon flux takes the form

$$F(\tau) \equiv \langle J_1(0)J_1(\tau)\rangle = (\delta Q\Gamma_1)^2 \{n_{10}^{+}[\overset{+}{G}{}^{10}_{00}(\tau)$$

$$+ 2\overset{+}{G}{}^{11}_{00}(\tau)] + 2n_{11}[\overset{+}{G}{}^{10}_{10}(\tau) + 2\overset{+}{G}{}^{11}_{10}(\tau)]\}, \tag{7}$$

where $\overset{+}{G}{}^{kl}_{ij}$ is the Green's function of the symmetrized system (1.18), (1.20), (1.23); see Discussion.

We will consider, for example, the term in Eq. (7) proportional to $\overset{+}{G}{}^{10}_{00}(\tau)$. This is expressed as the product of the following probabilities: The probability of being in the initial state (1,0) at initial time (the factor n_{10}^{+}); the probability of radiating a recordable photon at $t = 0$ ($\delta Q\Gamma_1$) after which the bichromophore ends up in the state (0,0); the probability of a transition by time $t = \tau$ to the state (1,0) [the probability is given by the Green's function $G^{10}_{00}(\tau)$]; the probability of photon radiation at final time ($\delta Q\Gamma_1$). The remaining contributions to Eq. (7) are analogous; these are the summation terms over the initial and intermediate states.

The structure of Eq. (7) is substantially different from population correlation function (4) due to the change in the bichromophore state as a result of photon emission. Function (7) is independent of the rates of processes (1.5), since energy exchange does not alter the fluorescence intensities. If we ignore cooperative processes (setting $\beta_{11} = 0$), Eq. (7) is consistent with the results from Ref. 3.

For simplicity we consider the case $r_0 = 0$, $\bar{\sigma}_1 = 0$ and we obtain from Eq. (7)

$$\frac{F(\tau)}{(\delta Q\Gamma_1)^2} = (2n_1)^2 + [2n_{11} - (2n_1)^2]\,\frac{e^{\lambda_+\tau} + e^{\lambda_-\tau}}{2}$$

$$- \Big\{(\Gamma_1 + \sigma_{10}I + \beta_{11})\Big[\frac{(n_{10}^{+})^2}{2} - n_{11}(n_{11} + n_{00}\Big]$$

$$+ 2n_1 n_{11}(\Gamma_1 + \sigma_{10}I)\Big\}\,\frac{e^{\lambda_+\tau} - e^{\lambda_-\tau}}{\lambda_+ - \lambda_-}, \tag{8}$$

where $\lambda_\pm$ are the eigenvalues of the system (1.18), (1.20), (1.23),

$$\lambda_\pm = -\frac{1}{2}\Big\{3(\Gamma_1 + \sigma_{10}I)$$

$$+ \beta_{11} \mp \sqrt{(\Gamma_1 + \sigma_{10}I)^2 + 2\beta_{11}\Big(\Gamma_1 - \sigma_{10}I + \frac{\beta_{11}}{2}\Big)}\Big\}. \tag{9}$$

Figure 2 gives the normalized autocorrelation functions $f(\tau)$ calculated consistent with Eqs. (6)–(8). It is clear that in all cases there is a negative correlation (photon antigrouping). Photon correlation is fundamentally different from the correlation of the populations of the radiating levels in this aspect (compare to Fig. 1 and its discussion).

With small times τ or at low intensities [Fig. 2(a)] the cooperative processes enhance antigrouping since they inhibit independent emission of two photons by the bichromophore monomers, and for successive emission of two pho-

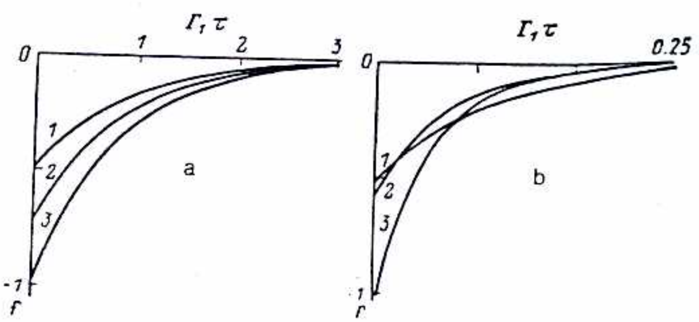

FIG. 2. Irreducible normalized correlation function $f(\tau)$ of fluorescence photon flux [Eqs. (6) and (8)]: (a) relative intensity of exciting radiation $I/I_1 = 0.1$, $\beta_{11}/\Gamma_1 = 0, 1, 100$ (curves 1–3); (b) $I/I_1 = 10$, $\beta_{11}/\Gamma_1 = 0, 10, 1000$ (curves 1–3).

tons a single monomer requires time (greater with large I) for reexcitation. In the case of finite saturation [Fig. 2(b)] the cooperative processes will suppress photon antigrouping with relatively large times since they reduce their correlation time.

Photon antigrouping (a negative correlation of Eqs. (6) and (8), see also Fig. 2) suggests that there is a dead time following photon emission and during this period the probability of emitting a second photon is reduced (compare to model experiments[4]). Such photon statistics are characterized by a dispersion that is smaller than for a Poisson distribution, and are called sub-Poisson statistics.

Let M be the number of photons striking the photodetector over the sampling time T. The dispersion $\langle \Delta M^2 \rangle$ is expressed through the autocorrelation function of photon flux (6) (see, for example, Ref. 5)

$$\langle \Delta M^2 \rangle = \langle M \rangle (1+\xi), \qquad \xi = 2\frac{\langle J_1 \rangle}{T}\int_0^T dt' \int_0^{T-t'} f(t)\, dt. \tag{10}$$

Since $f(\tau) < 0$ (antigrouping) it is obvious that $\xi < 0$, i.e., the dispersion of the number of photons is in fact less than for a Poisson flux. The corresponding parameter for the photosamples is obtained by multiplying ξ [EQs. (10)] by the quantum efficiency of the photocathode.

Assuming $T \gg \Gamma_1^{-1}$ we find $\xi = 4\delta Q \Gamma_1 n_1 \int_\infty^0 f(t)dt$. Using Eqs. (9), (10), and (6) we obtain a closed expression for the parameter ξ through the average populations (1.24)–(1.28)

$$\xi = \frac{\delta Q \Gamma_1}{2n_1[2(\Gamma_1 + \sigma_{10} I)^2 + \beta_{11}(\Gamma_1 + 2\sigma_{10} I)]}$$

$$\times \Big\{ (n_{11} - 2n_1^2)[3(\Gamma_1 + \sigma_{10} I) + \beta_{11}]$$

$$- (\Gamma_1 + \sigma_{10} I + \beta_{11}) \left[\frac{(n_{10}^{+})^2}{2} - n_{11}(n_{00} + n_{11}) \right] - 2n_1 n_{11}(\Gamma_1 + \sigma_{10} I) \Big\}. \tag{11}$$

Figure 3 illustrates the behavior of ξ as a function of saturation at various rates β_{11}. It is clear that at low intensities the cooperative processes will cause a substantial relative growth of ξ (in this case, however, $|\xi|$ is not large). The modulo value of ξ increases with increasing I, reaching a maximum at finite values of I that are lower the greater β_{11}. The maximum value of $|\xi|$ is one-half in the absence of cooperative processes ($\beta_{11} = 0$); it tends to 1/2 also in the case of very weak cooperation ($\beta_{11} \gg \Gamma_1$) and is somewhat less than 1/2 with intermediate values of β_{11}. At high intensities $|\xi|$ approaches zero and cooperative processes diminish $|\xi|$, i.e., the photon flux is Poissonized.

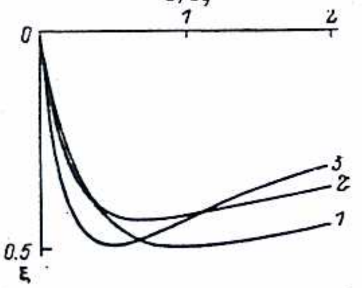

FIG. 3. Parameter ξ of the fluorescence sub-Poisson statistics plotted as a function of the relative intensity of exciting radiation. $\beta_{11}/\Gamma_1 = 0, 1, 100$ (curves 1–3).

Reference 6 has suggested that cooperative quenching in a chromophore system will always result in more clearly expressed sub-Poisson statistics (increasing $|\xi|$). These results shows that this proposition is valid for bichromophores only at very low radiation intensities. We note that Ref. 6 employed a square-law approximation equivalent to ignoring the population correlation of the monomers. Of course this approximation is more realistic for a system of many chromophores rather than for a bichromophore.

DISCUSSION

We will briefly summarize the primary principles and results from the present study.

The stationary fluctuations in the populations and the fluorescence photon flux are characterized by paired correlation functions [See Eqs. (4), (5) and (6)–(8) as well as Figs. 1–3]. Photon correlation has an antigrouping character; it is also responsible for their sub-Poisson statistics. The correlation function (6) of fluorescence photons and the dispersion (10) of their number are directly measured by photon-correlation spectroscopy (see, for example, Ref. 2 and 7). These bear information on the rate constant of cooperative processes and on the relaxation constants of the chromophores.

The population correlation of the monomers does not reduce to the photon correlation [compare Eqs. (4) and (7)] since the emission process alters the bichromophore state. It is clear that the population correlation can be measured only by means of weakly perturbing effects. Such effects include quasielastic scattering of test particles such as photons, neutrons, or slow electrons. The autocorrelation function of the scattered particle flux is determined by the dynamic formfactor $S_{\mathbf{q}}(t) = \langle \rho_{\mathbf{q}}(t)\rho_{\mathbf{q}}^*(0) \rangle$, where $\rho_{\mathbf{q}}(t)$ is the Fourier transform of the scatterer density $\rho(\mathbf{r},t)$ at time t (inferring the density interacting with the scattered particle); $\mathbf{q}$ is the change in the wave vector upon scattering. The energy distribution of the scattered particles is given by the Fourier transform in t of this same form factor.

We calculate $S_{\mathbf{q}}(t)$ for a symmetric bichromophore

$$S_{\mathbf{q}}(t) = 2|\bar{\rho}_{\mathbf{q}}|^2(1 + \cos \mathbf{q}\cdot\mathbf{R})$$

$$+ 2|(\rho_1 - \rho_0)_{\mathbf{q}}|^2[\bar{C}(t) + C(t)\cos \mathbf{q}\cdot\mathbf{R}], \tag{12}$$

where $\mathbf{R}$ is the radius-vector separating the monomers in the bichromophore; ρ_0 and ρ_1 are the monomer densities in states S_0 and S_1, respectively; $\bar{\rho} = n_0\rho_0 + n_1\rho_1$. It is clear that the dynamic form factor of the bichromophore is in fact expressed through the correlation functions (5) and bears information on energy exchange constants (1.5).

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[1)] Throughout we will use the notation of Ref. 1. The designation Eq. (1.25) means Eq. (25) in Ref. 1, etc.

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Four-photon spectroscopy of GaSe: methods of temporal analysis

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Results of spectroscopic studies of GaSe (293 K) in a range covering the regions of transparency and of single-photon interband and exciton transitions are presented. A set of nonlinear spectroscopy methods such as the test beam method and saturation spectroscopy are used to identify the mechanisms of nonlinearity and characteristic relaxation processes.

INTRODUCTION

Semiconductors and, in particular, GaSe constitute a complex subject for spectroscopic studies, since the response of the medium is governed by several nonlinearity mechanisms, each of them is characterized by its own type of relaxation processes with the corresponding times, there is no universal theoretical model describing the results obtained by different spectroscopic methods under different conditions, and the experimental data are simultaneously affected by a number of factors that are difficult to take into account (dependence of the transmittance of the sample on the intensities, frequencies of the interacting waves, etc.).

The characteristic times of the relaxation processes are measured by comparing the experimental data with specific theoretical models.[1–5] Therefore, such models significantly determine the conclusions reached by the authors. Generally, the experimenters use the simplest models of a given selected (sometimes unjustifiably) relaxation process. The erection of a more complete theory is complicated by the fact that the accumulated experimental material actually consists of a set of expertimental data obtained under different conditions which are not always clearly specified. The physics of the processes taking place can only be determined through the comprehensive application of the maximum possible number of spectroscopic methods under the same conditions. Combined analysis of the experimental data obtained can yield new information, not attainable by each of the methods taken separately, on the mechanisms of relaxation processes. Reference 6 substantiated the need for such an approach and indicated ways of implementing it.

The present paper includes the results of spectroscopic studies of GaSe (293 K) in a range covering the regions of transparency and of single-photon interband and exciton transitions. The utilization of the capabilities of the experimental arrangement, which was analogous to that described in Ref. 6, permitted an effective application of a set of methods of nonlinear spectroscopy (NS) such as the test-beam (TB) method and saturation spectroscopy (SS) to the identification of nonlinearity mechanisms and characteristic relaxation processes.

The results obtained by use of methods of degenerate four-photon spectroscopy (DFPS) and biharmonic pumping (BP) will be published separately.

APPLICATION OF METHODS OF NONLINEAR SPECTROSCOPY

The NS methods are based on the process of generation of a wave of nonlinear polarization with frequency $\omega_4 = \omega_l - \omega_m + \omega_n$, also lying in the range studied, and wave vector $\mathbf{K}_4 = {}_l + \mathbf{K}_n - \mathbf{K}_m$ $(l,m,n = 1,2,3)$. During the experiments, depending on the NS method, we recorded the energy of the corresponding spatial component (fixed direction of $\mathbf{K}_4$) of the field, and controlled the times of arrival, polarization, frequencies, and intensities of the pulses interacting in the sample.

Each of the NS methods investigates the dependence of nonlinear response on certain selected variables which determine the conditions of excitation or probing of the sample. The classification of the methods is based on their subdivision into dynamic and quasistationary ones. The former involve temporal analysis of the relaxation of nonlinear response. Their resolution is determined by the duration of the pulses employed (in practice ~25 psec) and is completely sufficient for the study of nanosecond and subnanosecond processes (for example, recombination and spatial diffusion of carriers in semiconductors). In the study of subpicosecond dynamics (polarization relaxation), the study of the frequency of dispersion of nonlinear susceptibility—spectral analysis of nonlinear response—turns out to be more promising. This analysis is carried out in the presence of a parametric interaction of waves of different frequencies that is stationary relative to fast relaxation processes.[5,6] The specif-

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