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Spectral diffusion induced by sudden jump processes

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Abstract

A concise analytical method is given to describe the spectral diffusion in the system with several sudden jump modulators from the frequency domain viewpoint. Spectral diffusion of chromophores in low temperature glasses is investigated by this method with the standard two-level system model. Under the standard approximation, the spectral diffusion has exactly the same form as the stimulated photon echo result. The time-dependent heterogeneous line width increases as the logarithm of waiting time. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Homogeneous spectral line width of guest chromophores is usually well described two phenomenological constants longitude relaxation time T_1 and transversal relaxation time T_2 ($(1/T_2) = (1/T_2') + (1/2T_1)$), while $1/2\pi T_1$ corresponds to the *natural line width* and T_2' is called the *pure dephasing time*. It does not change with measurement time. But, if the guest chromophores undergo stochastic frequency modulations induced by the host environment, it will cause a additional spectral broadening. Random frequency modulations exist in many systems, such as magnetic system [1] and glasses at low temperature [2], dye solution [3], in which the stochastic perturbations

arise from electronic/nuclear spins, random array of two-level systems (TLSs) and phonon bath, respectively. Because the interactions have various time scales, they can lead to a time-dependent broadening spectral line (heterogeneous line), the process is so-called spectral diffusion (SD). SD is usually investigated by hole-burning and stimulated photon echo experiments by varying different measurement time parameters T_W (e.g., time separation and waiting time in hole-burning and stimulated echo experiments, respectively). Characterizing SD can give us a complete picture of the microscopic dynamics of the host system and its influence on the guest chromophore.

Spectral diffusion is a typical conception in frequency domain, but almost no theoretical work started from the *pure* frequency domain point. In frequency domain case, the theoretical treatment is rather difficult even for the system modulated by a single random process, because the radiation field

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couples with the system in the kinetic equations for the hole-burning process [4]. When the number of the stochastic processes increases, the equations become more and more complex [5]. For a practical system containing an infinity of modulators, it is impossible to give a clear form expressing the SD process. So, hole-burning is considered as equivalent to the stimulated echo, which is related to the stimulated echo decay function through a Fourier transform [6–8]. In fact, many experimental results have shown the difference in the behavior between the frequency and time domains. For example, in ruby at low temperature, the line width measured by free induced decay (time domain) is 10 times large than that by transient hole-burning (frequency domain) [9,10]; in some low temperature glasses, SD processes measured by stimulated photon echo and hole-burning also have different behaviors [11,12]. So, it is necessary to investigate SD in the pure frequency domain.

Computer simulation seems to promise an opportunity. For example, Monte Carlo simulations have been applied to study the time-dependent broadening processes in $\text{Pr}^{3+}:\text{YAlO}_3$ (time domain) [13] and $\text{Eu}^{3+}:\text{YAlO}_3$ (frequency domain) [14]; single molecule spectroscopy in disordered systems has also been simulated by the random walk method [15]. In fact, all these works did not consider the radiation field coupling, and were simulated under very simple models. For much more complex systems, such as low temperature glasses, the average over the parameters of the random array of TLSs is rather difficult to carry out even without considering the numerous modulators in the host. To give a clear picture of SD in the frequency domain, a concise analytical method is needed.

In this paper, we only consider the SD process after switching off the radiation field; first we develop a concise analytical method to describe the SD in system with several sudden jump processes. Then, starting from the standard TLSs model, we study the SD in low temperature glasses for pure frequency domain case. Under the standard approximation, the result of SD goes as exactly the same form for time domain case.

2. Spectral diffusion for systems modulated by $2n$ sudden jump processes

We consider a system is modulated by $2n$ independent sudden jump modulators, which each flips between the energy states of $-E$ and E and causes $\pm\delta$ frequency shift of the system. The sum of frequency shift ω , spreads within the range $[-2n\delta, 2n\delta]$. Assume among $2n$ modulators $n+k$ with E and $n-k$ with $-E$, hence $\omega = 2k\delta$. The stationary distribution density at $\omega = 2k\delta$ is

$$P(k) = \frac{(2n)!}{(n+k)!(n-k)!} \left(\frac{1}{2}\right)^{2n} \quad (1)$$

If n is large, it can be approximated by a Gaussian form

$$P(k) = \sqrt{\frac{1}{n\pi}} e^{-k^2/n} \quad (2)$$

We denote $P_{if}(t)$ as the jump probabilities in the time interval of t , for the path where i is the initial state, f is the final state ($if = \pm 1$), and can be given by

$$\begin{aligned} P_{+-}(t) &= \rho_{\text{eq}-}(1 - e^{-Rt}), \\ P_{-+}(t) &= \rho_{\text{eq}+}(1 - e^{-Rt}), \\ P_{++}(t) &= 1 - P_{+-}(t), \\ P_{--}(t) &= 1 - P_{-+}(t) \end{aligned} \quad (3)$$

where $\rho_{\text{eq}\pm}$ are the equilibrium population densities of up and lower states, respectively, and R is the sum of the upward and downward rate constants. According to the detailed balance at every temperature, we have $\rho_{\text{eq}+}/\rho_{\text{eq}-} = \exp(-2\beta E)$, where $2E$ is the energy difference between the up and lower levels of the modulator.

If j is out of $n+k$ modulators initially with E , and $(j-i)$ is out of $n-k$ modulators initially with $-E$ flip odd times within time t , $\omega(t) = 2(k-i)\delta$. The probability is

$$\begin{aligned} P(i, t) &= \sum_{j=\max(0, i)}^{\min(n+k, n-k+i)} \frac{(n+k)!(n-k)!}{j!(n+k-j)!(j-i)!(n-k-j+i)!} \\ &\quad \times P_{+-}^j P_{-+}^{j-i} (1 - P_{+-})^{n+k-j} (1 - P_{-+})^{n-k-j+i} \end{aligned} \quad (4)$$

where $k-n \leq i \leq k+n$.

Time dependencies of the line width can be obtained from the first and the second moments of the distribution equation (4):

$$\begin{aligned}
 \langle i \rangle &= \sum_{i=-(n-k)}^{n+k} \sum_{j=\max(0,i)}^{\min(n+k,n-k+i)} \frac{i(n+k)!(n-k)!}{j!(n+k-j)!(j-i)!(n-k-j+i)!} \\
 &\quad \times P_{+-}^j P_{-+}^{j-i} (1-P_{+-})^{n+k-j} (1-P_{-+})^{n-k-j+i} \\
 &= \sum_{i=0}^{n-k} \sum_{j=0}^{n+k} \frac{(j-i)(n+k)!(n-k)!}{j!(n+k-j)!i!(n-k-i)!} \\
 &\quad \times P_{+-}^j P_{-+}^i (1-P_{+-})^{n+k-j} (1-P_{-+})^{n-k-i} \\
 &= \sum_{j=1}^{n+k} \frac{(n+k)!}{(j-1)!(n+k-j)!} P_{+-}^j (1-P_{+-})^{n+k-j} \\
 &\quad - \sum_{i=1}^{n-k} \frac{(n-k)!}{(i-1)!(n-k-i)!} P_{-+}^i (1-P_{-+})^{n-k-i} \\
 &= (n+k)P_{+-} - (n-k)P_{-+} \\
 &= (1-e^{-Rt})[(n+k)\rho_{\text{eq-}} - (n-k)\rho_{\text{eq+}}]
 \end{aligned} \tag{5}$$

$$\begin{aligned}
 \langle i^2 \rangle &= \sum_{i=-(n-k)}^{n+k} \sum_{j=\max(0,i)}^{\min(n+k,n-k+i)} \frac{i^2(n+k)!(n-k)!}{j!(n+k-j)!(j-i)!(n-k-j+i)!} \\
 &\quad \times P_{+-}^j P_{-+}^{j-i} (1-P_{+-})^{n+k-j} (1-P_{-+})^{n-k-j+i} \\
 &= \sum_{i=0}^{n-k} \sum_{j=0}^{n+k} \frac{(j-i)^2(n+k)!(n-k)!}{j!(n+k-j)!i!(n-k-i)!} \\
 &\quad \times P_{+-}^j P_{-+}^i (1-P_{+-})^{n+k-j} (1-P_{-+})^{n-k-i} \\
 &= [(n+k)P_{+-} - (n-k)P_{-+}]^2 \\
 &\quad + [(n+k)P_{+-}(1-P_{+-}) - (n-k)P_{-+}(1-P_{-+})]
 \end{aligned} \tag{6}$$

and we have

$$(\langle i^2 \rangle - \langle i \rangle^2)^{1/2} = [(n+k)P_{+-}(1-P_{+-}) - (n-k)P_{-+}(1-P_{-+})]^{1/2} \tag{7}$$

Since the system is always in the state of dynamical equilibrium, approximately, $n+k$ and $n-k$ are equal to $2n\rho_{\text{eq+}}$ and $2n\rho_{\text{eq-}}$, respectively. Hence, $\langle i \rangle$ is always zero. Note $\rho_{\text{eq+}} + \rho_{\text{eq-}} = 1$, Eq. (7) can be expressed as

$$(\langle i^2 \rangle - \langle i \rangle^2)^{1/2} = [2n \operatorname{sech}^2(\beta E)(1 - e^{-2Rt})]^{1/2} \tag{8}$$

The time-dependent heterogeneous line width is

$$H(t) = H(\infty) \operatorname{sech}(\beta E)(1 - e^{-2Rt})^{1/2} \tag{9}$$

From Eq. (2), for large n as t approaches to infinity, the line shape is a Gaussian one and $H(\infty)$ is $(2n)^{1/2}\delta$.

When temperature is high enough, $\operatorname{sech}(\beta E) \approx 1$. The sudden jump process reduces to the bivalued random telegraph one ($P_{+-} = P_{-+}$). The spectral diffusion goes as $H(t) = H(\infty)(1 - e^{-4Wt})^{1/2}$, where W is jump rate of random telegraph process.

As we have motioned above, $\langle i \rangle$ is always zero if we consider the system is in the state of dynamical equilibrium. That means the peak of the spectral line will never shift during the spectral diffusion. One can find that this treatment also implies the condition of resonant laser exciting. In the case of arbitrary exciting frequency (different value of k), the peak shift of heterogeneous line can be given by

$$\begin{aligned}
 \omega_0(t) &= 2\delta(k - \langle i \rangle) \\
 &= 2\delta\{k - (1 - e^{-Rt})[(n+k)\rho_{\text{eq-}} - (n-k)\rho_{\text{eq+}}]\}
 \end{aligned} \tag{10}$$

For random telegraph process, we have

$$\omega_0(t) = 2\delta(k - \langle i \rangle) = 2k\delta e^{-2Wt} \tag{11}$$

3. Dephasing mechanism in low temperature glasses

In low temperature glasses, SD has been extensively studied by both experimental and theoretical methods [6–8,11,16]. SD can be observed in a very large time range from nanoseconds up to days [16].

It is well known that low temperature glass can be described as a random array of TLSs, which was first proposed by Phillips to explain the acoustic properties of disorder systems [17]. The Hamiltonian of the TLS is determined by two parameters A , J [18]. Here, A is the asymmetry, i.e., the energy gap between the localized states; and J is the tunneling matrix element that couples the two localized states. These TLSs are coupled to the acoustic phonons of the glassy host. The coupling induces flip-flop between the two eigenstates of the TLS, which is called phonon-assisted tunneling. The sum of the upwards and downwards jump rate can be given by Fermi's golden rule [19]

$$R = cJ^2 E \coth(\beta T/2) \tag{12}$$

where $E = (A^2 + J^2)^{1/2}$ is the energy gap between the TLS eigenstates, c is a TLS–phonon coupling constant, and $\beta = 1/k_B T$ (T is temperature, and k_B is Boltzmann's constant).

The sudden jump model is used to describe the TLS–chromophore interaction, which was first introduced to explain the relaxation process in the magnetic system at low temperature [20,21]. This model promises a unique opportunity to solve the coherent transient problems analytically. For a single TLS at the position r , the flip-flop leads to a shift in the chromophore's transition frequency by $\pm\omega(r)$. $\omega(r) = \alpha(A/2Er^3)$, where α is the TLS–chromophore interaction constant, and r is the distance between TLS and chromophore. The probability distributions of the parameters A, J are given by [22,23]

$$P_A(A) = \frac{1 + \mu}{A_{\max}^{1+\mu}} A^\mu, \quad 0 \leq A \leq A_{\max}, \quad (13)$$

$$P_J(J) = \frac{1}{J \ln(J_{\max}/J_{\min})}, \quad 0 \leq J \leq J_{\max}$$

where μ is the phenomenological exponent for the distribution of energy asymmetries. The above model is called the *standard model* for low temperature glasses, it leads to a approximately linear relation of $\ln(T_W)$ for SD [7,22]. In theoretical treatments, the value of μ is taken in the range from 0 to 1/3. It is widely believed that $\mu = 1/3$ is more suitable for many glasses. The upper cutoffs are taken as $J_{\max}, A_{\max} \gg k_B T$. The low cutoff J_{\min} is taken as $J_{\min}^2 \ll 2ckT\tau_{\text{exp}}$, where τ_{exp} is the longest imaginable waiting time (on the order of 10^6 s [16]). It will be essentially a parameter-independent model, if one choose the cutoff parameters in this way.

Using the relation $A = \{E^2 - R/[cE \coth(\beta E/2)]\}^{1/2}$, the (A, J) average can be transformed into a convenient (E, R) average. If a function $f(E, R)$ is essentially zero when $E > J_{\max}, A_{\max}$, the (A, J) average of $f(E, R)$ can be expressed as [22,23]

$$\langle f(E, R) \rangle = \frac{P_0}{2} (kT)^{1+\mu} \int_{E_{\min}}^{E_{\max}} dE E^\mu \int_{R_{\min}(E)}^{R_{\max}(E)} \frac{dR}{R} \times \left(1 - \frac{R}{R_{\max}(E)}\right)^{(\mu-1)/2} f(E, R) \quad (14)$$

where

$$\begin{aligned} P_0 &= \frac{1 + \mu}{A_{\max}^{1+\mu} \ln(J_{\max}/J_{\min})}, \\ R_{\max}(E) &= cE^3 \coth(\beta E/2), \\ R_{\min}(E) &= cE J_{\min}^2 \coth(\beta E/2), \\ E_{\min} &= J_{\min} \quad \text{and} \quad E_{\max} = \sqrt{(A_{\max}^2 + J_{\max}^2)} \end{aligned} \quad (15)$$

Although the standard model is very successful, there still some experiments cannot be explained. For this purpose, Silbey et al. modified the model by introducing another phenomenological factor v , while -1 the exponent for the distribution of the tunneling matrix element was replaced by $-(1 - v)$ [23]. This model gave the time behavior of SD as $1 - \exp[-v/2 \ln(R_{\text{eff}} T_W)]$, where R_{eff} was the effective maximum averaged over the energy gap E . There were also other modified models, which started from various distribution of the parameters A and J . As Bai and Fayer have pointed, the different SD behavior directly result from the different flip rate distribution $P(R)$ [24]. They derived a general relation between a T_W dependence measurement and $P(R)$, which the derivative of the stimulated echo decay function is directly proportional to the Laplace transform of $P(R)$.

Beside the flip rate distribution $P(R)$, other conditions can also effect the SD process. Recently, Lazonder et al. proved that, the optical excitation-induced heating mainly caused the discrepancy from the predication by the standard model [25]. Considering the time dependent heating, they demonstrated that it was no longer necessary to extend the standard model by means of adjustable parameters of $P(R)$.

Since the standard model is widely adopted, for the purpose of comparison, we will use it in this paper.

4. Spectral diffusion by frequency domain method

To investigate SD in low temperature glasses, we treat each subset with modulation frequency $\omega + \Delta\omega$ by the above model, then need finish the averages of the spatial and the parameters of the TLS. For a single TLS, the total heterogeneous

line width is the sum of all subsets, which is the average over the TLS–chromophore distances and orientations. Considering continuous limit, we have

$$\begin{aligned}
 H^2(T_W) &= 4\pi(1 - e^{-2RT_W})\operatorname{sech}^2(\beta E/2) \\
 &\quad \times \int_{r_{\min}}^{\infty} \omega^2 \rho r^2 dr \\
 &= H^2(\infty)\operatorname{sech}^2(\beta E/2)(1 - e^{-2RT_W}), \\
 \left(H(\infty) &= \sqrt{\frac{2\pi\alpha\rho A\omega_{\max}}{3E}} \right) \quad (16)
 \end{aligned}$$

where ρ is the TLS density, $\omega_{\max} = (\alpha A/2Er_{\min}^3)$, and r_{\min} is minimum distance. Note that the lower limit of the integral cannot be extended to zero, in contrast to time domain case. One can find that the static broadening $H(\infty)$ is just the second moment line width in the continuous lattice limit, which is extensively used in magnetic systems [26].

If we assume r_{\min} is equal to the distance between the two nearest TLSs, using the relation $\rho r_{\min}^3 = 0.17$ [27], we get

$$H(E, R, T_W) = 1.4\pi^{1/2}\alpha\rho\frac{A}{E}\operatorname{sech}(\beta E/2)(1 - e^{-2RT_W})^{1/2} \quad (17)$$

In following, we take the average over the parameters E, R by both two paths, respectively. Because of the $\operatorname{sech}(\beta E/2)$ factor, $H(E, R, T_W)$ is obviously the required class of functions averaging over E, R . Using Eqs. (14) and (15), set $x = 2RT_W$, $z = \beta T$, we have

$$\begin{aligned}
 H(T_W) &= 0.7\pi^{1/2}\alpha P_0\rho(k_B T)^{1+\mu} \\
 &\quad \times \int_{\beta E_{\min}}^{\beta E_{\max}} dz z^\mu \operatorname{sech}(z/2) \\
 &\quad \times \int_{2\tilde{R}_{\min}(z)T_W}^{2\tilde{R}_{\max}(z)T_W} \frac{dx}{x} \left(1 - \frac{x}{2\tilde{R}_{\max}(z)T_W} \right)^{\mu/2} \\
 &\quad \times (1 - e^{-x})^{1/2} \quad (18)
 \end{aligned}$$

where $\tilde{R}_{\max}(z) = \Gamma z^3 \coth(z/2)$, $\tilde{R}_{\min}(z) = \Gamma z J_{\min}^2 \times \coth(z/2)$, $\Gamma = c(k_B T)^3$.

By the same consideration as Ref. [22], the lower and up limits of the z integral can be safely taken to 0 and infinity, respectively. Then consider the lower limit of the x integral. With the assumption of J_{\min} , one has $2ck_B T \tau_{\exp} J_{\min}^2 T_W \ll T_W/\tau_{\exp}$. Since the

largest waiting time $T_W \sim \tau_{\exp}$, so $2\tilde{R}_{\min}(z)T_W \ll 1$ is never invalid. We can also set it to zero. These are so called the *standard approximation* for SD [22]. With these treatment, Eq. (18) can be given as

$$\begin{aligned}
 H(T_W) &= 0.7\pi^{1/2}\alpha P_0\rho(k_B T)^{1+\mu} \\
 &\quad \times \int_0^{\infty} dz z^\mu \operatorname{sech}(z/2) \\
 &\quad \times \int_0^{2\Gamma T_W z^3 \coth(z/2)} \frac{dx}{x} \\
 &\quad \times \left(1 - \frac{x}{2\Gamma z^3 \coth(z/2)T_W} \right)^{\mu/2} (1 - e^{-x})^{1/2} \quad (19)
 \end{aligned}$$

Furthermore, one can also substitute the term $\left(1 - (x/2\tilde{R}_{\max}(z)T_W) \right)^{\mu/2}$ by 1, which is a very well approximation except x is close to the upper limit of the x integral and T_W has a very small value. For the x integral, we can divide it into two parts

$$\begin{aligned}
 &\int_0^{2\Gamma T_W z^3 \coth(z/2)} \frac{dx}{x} (1 - e^{-x})^{1/2} \\
 &= \int_0^1 \frac{dx}{x} (1 - e^{-x})^{1/2} + \int_1^{2\Gamma T_W z^3 \coth(z/2)} dx \\
 &\quad \times \left\{ \frac{1}{x} - \frac{1}{x} [1 - (1 - e^{-x})^{1/2}] \right\} \\
 &= 1.852 + \ln[2\Gamma z^3 \coth(z/2)T_W] \\
 &\quad + \int_1^{2\Gamma T_W z^3 \coth(z/2)} dx \frac{1}{x} [(1 - e^{-x})^{1/2} - 1] \quad (20)
 \end{aligned}$$

The last term rapidly tends to be a constant as the up limit of the integral increases. Since $2\Gamma T_W z^3 \coth(z/2)$ usually has a large value, we can substitute it with infinity. The value of the numerical integral is -0.117 , this small value promises that the above approximation is valid for $2\Gamma T_W z^3 \coth(z/2) > 1$. Then we have

$$\begin{aligned}
 H(T_W) &= 0.7\pi^{1/2}\alpha P_0\rho(k_B T)^{1+\mu} \\
 &\quad \times \{ C(\mu) \ln[c(k_B T)^3 T_W] + D_1(\mu) \} \quad (21)
 \end{aligned}$$

where

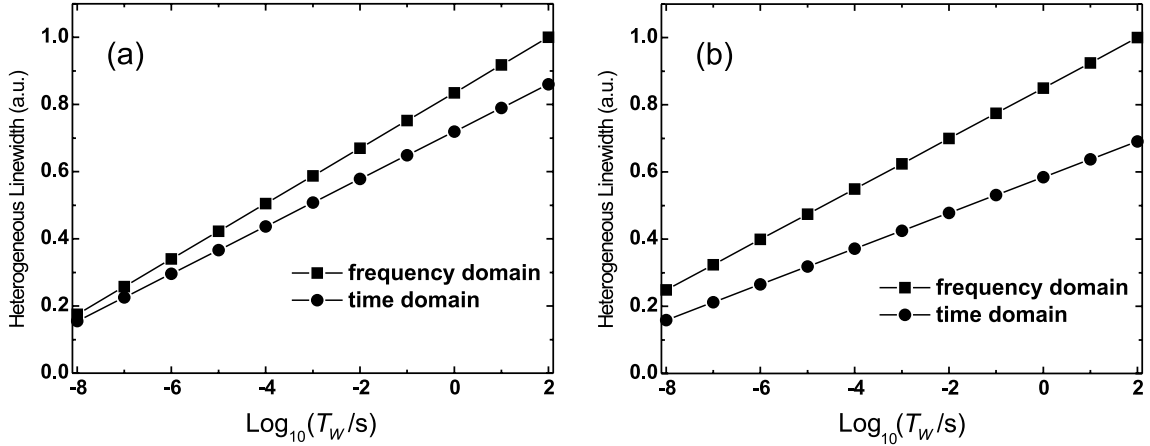


Fig. 1. Waiting time dependence of heterogeneous line width, results in frequency and time domains under the standard approximation: (a) $\mu = 0$; (b) $\mu = 1/3$.

$$C(\mu) = \int_0^{\infty} dz z^{\mu} \operatorname{sech}(z/2)$$

$$D_1(\mu) = 2.428C(\mu) + \int_0^{\infty} dz z^{\mu} \operatorname{sech}(z/2) \quad (22)$$

$$\times \ln[z^3 \coth(z/2)]$$

Note that the above heterogeneous line width contains all broadening contributions except for the natural line width. Hence, for the pure SD line width, one need to subtract the pure dephasing line width of two-pulse echo decay ($1/\pi T_2'$). In fact, this procedure should be done at the beginning of the average over E , R . But it is difficult to determinate T_2' for a single TLS, so is convenient to subtract the averaging $1/\pi T_2'$ from Eq. (21). Under the standard approximation, $1/\pi T_2'$ is well known as [7,22]

$$\frac{1}{\pi T_2'} = \frac{\pi^2}{6} \alpha P_0 \rho (k_B T)^{1+\mu} B(\mu) \quad (23)$$

where $B(0) = 7.328$, and $B(1/3) = 7.881$.

Subtracting this contribution, we get the final SD line width for the *pure* frequency domain case

$$H(T_W) = 0.7\pi^{1/2} \alpha P_0 \rho (k_B T)^{1+\mu} \times \{C(\mu) \ln[c(k_B T)^3 T_W] + D(\mu)\} \quad (24)$$

where $D(\mu) = D_1(\mu) - 0.238\pi^{3/2} B(\mu)$. $C(\mu)$ and $D(\mu)$ can be calculated numerically. For example, $C(0) = \pi$, $C(1/3) = 3.788$, and $D(0) = 3.498$, $D(1/3) = 8.766$.

It is very amazing that the result is exactly the same as the time domain one (the stimulated echo decay result) under the standard approximation (see Eq. (36) in Ref. [22]), however the values of $C(\mu)$ and $D(\mu)$ are different. Since we already assume that T_W is not very small, the above result does not give a zero value when $T_W \rightarrow 0$. For sufficiently large T_W , the factor $D(\mu)$ can be neglected. SD is dominated by the term $\ln[c(k_B T)^3 T_W]$. The time-dependent heterogeneous line width is proportional to $T^{1+\mu}$ with a multiplicative $\ln T$ correction, and increases as $\ln T_W$. Note that $C(0) = 2$ and $C(1/3) = 2.042$ for time domain case, we have the ratios of the SD line width in two domains are 1.2 and 1.4 for $\mu = 0$, $1/3$, respectively. The frequency method gives a little larger value of the SD line width. In Fig. 1, we show the comparison of the dependence of the heterogeneous line width $H(T_W)$ on T_W over 11 orders, for frequency domain and time domain under the standard approximation. In the calculation, we use the typical values of the parameters $ck_B^3 = 10^9 \text{ K}^{-3} \text{ s}^{-1}$, $T = 1 \text{ K}$.

5. Conclusions

We develop a concise analytical method to describe the spectral diffusion in the system with several sudden jump modulators from frequency

domain point. Spectral diffusion of chromophores in low temperature glasses is investigated by this method with the standard TLS model. Under the standard approximation, the spectral diffusion has exactly the same form as the stimulated photon echo result, however it gives a little larger value of the line width. The time-dependent heterogeneous line width is proportional to $T^{1+\mu}$ with a multiplicative $\ln T$ correction, and increases as $\ln T_W$.

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References

- [1] M. Macfarlane, R.M. Shelby, in: A. Kaplyanskii, R.M. Macfarlane (Eds.), *Spectroscopy of Solids Containing Rare-Earth Ions*, Elsevier Science, North-Holland, Amsterdam, 1987 (Chapter 3).
- [2] W.A. Phillips, *Rep. Prog. Phys.* 50 (1987) 1657.
- [3] J.-Y. Bigot, M.T. Portella, R.W. Schoenlein, C.J. Bardeen, A. Migus, C.V. Shank, *Phys. Rev. Lett.* 66 (1991) 1138.
- [4] H. Wang, S. Huang, *Opt. Commun.* 162 (1999) 148.
- [5] S.Ya. Kilin, A.P. Nizovtsev, P.R. Berman, C. Von Borzyskowski, J. Wrachtrup, *Phys. Rev. B* 58 (1998) 8997.
- [6] M. Berg, C.A. Walsh, L.R. Narasimhan, K.A. Littau, M.D. Fayer, *J. Chem. Phys.* 88 (1988) 1564.
- [7] Y.S. Bai, M.D. Fayer, *Comments Condens. Matt. Phys.* 14 (1989) 343.
- [8] A. Suárez, R. Silbey, *Chem. Phys. Lett.* 218 (1994) 445.
- [9] A. Szabo, T. Muramoto, R. Kaarli, in: J.H. Eberly, L. Mandel, E. Wolf (Eds.), *Coherent and Quantum Optics VI*, Plenum, New York, 1990, p. 1131.
- [10] A. Szabo, R. Kaarli, *Phys. Rev. B* 44 (1991) 12307.
- [11] D. Zimdars, M.D. Fayer, *J. Chem. Phys.* 104 (1996) 3865.
- [12] S.J. Zilker, L. Kador, J. Friebe, Yu.G. Vainer, M.A. Kol'chenko, R.I. Personov, *J. Chem. Phys.* 109 (1998) 6780.
- [13] Y.S. Bai, R. Kachru, *Phys. Rev. A* 44 (1991) R6990.
- [14] R. Yano, M. Mitsunaga, U. Uesugi, *Phys. Rev. B* 45 (1992) 12752.
- [15] Y. Tanimura, H. Takano, J. Klafter, *J. Chem. Phys.* 108 (1998) 1851.
- [16] H. Maier, B.M. Kharlamov, D. Haarer, *Phys. Rev. Lett.* 76 (1996) 2085.
- [17] W.A. Phillips, *J. Low. Temp. Phys.* 7 (1972) 351.
- [18] J.L. Skinner, H.P. Trommsdorf, *J. Chem. Phys.* 89 (1988) 897.
- [19] J. Jackle, *Z. Phys.* 257 (1972) 212.
- [20] P. Hu, S.R. Hartmann, *Phys. Rev. B* 9 (1974) 1.
- [21] P. Hu, L.R. Walker, *Phys. Rev. B* 18 (1978) 1300.
- [22] E. Geva, J.L. Skinner, *J. Chem. Phys.* 107 (1996) 7630.
- [23] R.J. Silbey, J.M.A. Koedijk, S. Volker, *J. Chem. Phys.* 105 (1996) 901.
- [24] Y.S. Bai, M.D. Fayer, *Phys. Rev. B* 39 (1989) 11066.
- [25] K. Lazonder, K. Duppen, D.A. Wiersma, *J. Phys. Chem.* 104 (2000) 6468.
- [26] A. Abragam, *Principles of Nuclear Magnetism*, Clarendon, Oxford, 1961.
- [27] S.K. Lyo, *Phys. Rev. B* 10 (1971) 3331.