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Long-time spectral diffusion induced by short-time energy transfer in doped glasses: concentration-, wavelength- and temperature dependence of spectral holes

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Abstract

We discovered a remarkable effect: spectral diffusion (SD) induced by energy transfer (ET), here called ET → SD. The ‘effective’ homogeneous linewidth Γ'_{hom} of the $S_1 \leftarrow S_0$ 0–0 band of free-base chlorin (H_2Ch) in polystyrene (PS) was followed as a function of concentration and excitation wavelength by hole-burning. The results cannot simply be explained by Förster’s ET mechanism: (1) the increase of Γ'_{hom} towards the blue is larger than expected and follows an S-shaped curve; (2) Γ'_{hom} increases linearly with concentration; and (3) the holes are Lorentzian. A model is proposed which predicts that Γ'_{hom} should increase with delay time, even at $T \to 0$. This is confirmed by experiments. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

To study the dynamics of glasses we doped them with a chromophore molecule as a probe. The ‘effective’ homogeneous linewidth Γ'_{hom} of the first electronic transition of the chromophore, when measured as a function of temperature T and the time scale t_d of the experiment, will give information on structural relaxation of the glass because of the coupling of the electronic transition of the guest with the environment.

Conventionally, the dynamic properties of glasses at low temperature are described in terms of low-frequency modes called two-level systems (TLSs) [1–4]. The TLSs are represented by double-well potentials with a broad distribution of barrier heights and asymmetries. A change in the glass structure is then modeled by transitions or ‘flips’ from one potential well to the other, covering a wide distribution of relaxation rates R [5–8].

In doped organic glasses at $T \leq 10$ K, it has been found that Γ'_{hom} varies with T and t_d as [9–13]

$$\Gamma'_{\mathrm{hom}} = \Gamma_0 + a(t_d) T^{1.3 \pm 0.1}, \quad (1)$$

where the residual linewidth Γ_0 for $T \to 0$ is given by $\Gamma_0 = (2\pi T_1)^{-1}$, with T_1 the excited-state lifetime. At low concentrations, $\Gamma_0 = (2\pi\tau_{\mathrm{fl}})^{-1}$, with t_{fl}

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the fluorescence lifetime of the probe molecule [11,12]. The second term in Eq. (1) accounts for two contributions, 'pure' dephasing (PD) which only depends on T and, what we will call here, 'normal' spectral diffusion (SD) attributed to the flipping of TLSs in the glass surrounding the excited molecule [11–16]. In a hole-burning (HB) experiment, SD manifests itself by an increase of Γ'_{hom} as a function of the delay t_{d} between burning and probing the hole. Note that for $T \to 0$ $\Gamma'_{\mathrm{hom}} \to \Gamma_0$. At concentrations higher than $c \approx 10^{-4}$ M, energy transfer (ET) between probe molecules may occur. The time a given molecule is in the excited state will then decrease because ET opens extra channels for relaxation. This leads to an increase of Γ'_{hom} through the first term of Eq. (1), which now becomes $\Gamma'_{\mathrm{hom}} = \Gamma_0 + \Gamma_0^{\mathrm{ET}}$, where $\Gamma_0^{\mathrm{ET}} = (2\pi\tau_{\mathrm{ET}})^{-1}$ is the ET rate.

Only two studies, to our knowledge, have been reported on the influence of concentration on spectral holewidths. Kulikov and Galaup [17] observed a broadening of the holes on the blue side of the 0–0 absorption band of chlorin and tetraphenylporphin in polystyrene (PS), which they attributed to ET without further analysis. Romanovskii et al. [18] also observed an increase of the holewidth with the energy of excitation in porphyrin-doped Langmuir–Blodgett films. They concluded that this effect could not be explained by Förster's ET mechanism, but did not analyze the problem further.

Thus far, it had not been realized that ET in glassy systems may trigger SD far in excess of that expected for a given transfer time τ_{ET}. By investigating the influence of 'downhill' ET on optical dephasing of the $S_1 \leftarrow S_0$ 0–0 band of free-base chlorin (H_2Ch) in PS by HB we also found that the results cannot simply be explained by Förster's ET mechanism, and looked for the process responsible for this discrepancy [19,20].

We first show in Section 3.1 by means of broadband fluorescence spectroscopy that ET occurs for H_2Ch in PS at $c \geq 5 \times 10^{-4}$ M. In Section 3.2 we present results on the shape of spectral holes at various concentrations, and on the dependence of Γ'_{hom} on concentration and excitation wavelength. In order to explain these results we propose a new mechanism: SD induced by ET, which we refer to as ET → SD. One of the predictions of this qualitative model is that Γ'_{hom} should increase with delay time, even at $T \to 0$. This expectation is confirmed by the experiments to be presented.

2. Experimental

2.1. Sample preparation

H_2Ch was prepared by reduction of free-base porphin (H_2P, Mad River) [21]. A few mg of H_2P were dissolved in ~50 ml pyridine under N_2 atmosphere, the solution was heated, and a few ml of hydrazine were added. To control the amount of H_2Ch formed, the maximum of the absorption band of H_2Ch in pyridine at 635 nm was monitored during the reduction and compared with the absorption bands of H_2P at 615 and 570 nm. The reaction was stopped when the latter bands had negligible intensity and the band of bacteriochlorin at 730 nm started to appear. This procedure took ~2 days. After the reduction, H_2Ch was purified through a 3 cm long Fluorisil column.

H_2Ch was mixed with a solution of PS (Aldrich, MW 280 000) in toluene. The solvent was evaporated by letting the solution dry for a few days. In this way transparent samples were obtained. The concentration c of the sample was determined from the optical density, $\mathrm{OD} = \varepsilon c d$, where ε is the extinction coefficient ($\varepsilon = 5 \times 10^4$ M^{-1} cm^{-1} at ~636 nm and $\varepsilon = 3.6 \times 10^3$ M^{-1} cm^{-1} at ~588 nm [22]) and d the thickness of the sample. The thickness of the films was determined with a micrometer. Absorption spectra at room temperature were obtained with a Perkin-Elmer spectrophotometer. The concentrations of the samples are given in Table 1. No sign of aggregation was observed in the absorption spectra up to concentrations of 6×10^{-3} M.

Table 1
Thickness d, optical density $\mathrm{OD} = \varepsilon c d$ at the maximum of the $S_1 \leftarrow S_0$ 0–0 band (~636 nm), and concentration c of the samples of H_2Ch in PS

d (mm)	OD_{636}	c (M)
2	0.1	1×10^{-5}
0.25	0.6	5×10^{-4}
0.02; 0.28	0.2; 2.8	2×10^{-3}
0.27	8.0	6×10^{-3}

The sample was placed in a home-built ^{4}He cryostat, of which the temperature was varied between 1.2 and 4.2 K by controlling the vapor pressure of the helium. The temperature was measured with a calibrated carbon resistor in contact with the sample with an accuracy better than 0.01 K [11,13].

2.2. Experimental apparatus for absorption-, fluorescence- and hole-burning spectroscopy

Broad-band absorption spectra were taken at 1.2 K by irradiating the sample with a halogen lamp. Its light beam was passed through a (7 cm thick) water filter to remove the infrared part of the spectrum and to avoid heating. The transmission was detected through the sample with a scanning 0.85 m double-monochromator (Spex 1402, resolution ~ 5 cm^{-1}) and a cooled photomultiplier (PM, EMI model 9658 R). Broad-band fluorescence spectra at 1.5 K were recorded by exciting the sample in the Soret band at 388 nm with a pulsed dye laser (Molectron DL 200, BBQ dye, bandwidth ~ 0.8 cm^{-1}, excitation power ~ 3 μJ/pulse) pumped by a nitrogen laser (Molectron UV 22, repetition rate ~ 20 Hz). The emission was detected with the scanning double-monochromator and the PM.

To determine the ‘effective’ homogeneous linewidth Γ'_{hom}, narrow holes were burnt using either a cw single-mode dye- or diode laser. The dye laser (Coherent 599-21, with intracavity assembly, $\Gamma_{\mathrm{laser}} \approx 2$ MHz, dye DCM), pumped by an Ar$^+$ laser (Spectra Physics 2030-15), was used for holes probed at long delay times after burning ($t_{\mathrm{d}} \approx 300$ s). Burning-fluence densities were used from Pt_{b}/A ~ 50 μJ/cm^2 to ~ 10 J/cm^2, depending on excitation wavelength and concentration. Burning times were ~ 1 min. The diode laser (Hitachi HL 6312G, current- and temperature-controlled, $\Gamma_{\mathrm{laser}} \approx 30$ MHz), operated at $\lambda = 633$ nm ($T_{\mathrm{laser}} \approx -5$°C) was used for (transient) holes probed at $t_{\mathrm{d}} \approx 100$ μs. Frequency-scan speeds up to 5 GHz/μs can be reached with this type of laser [12]. Burning-fluence densities from Pt_{b}/A ~ 20 nJ/cm^2 to ~ 4 μJ/cm^2 were used, with a burning time $t_{\mathrm{b}} \approx 100$ μs and laser powers between P ~ 5 μW and ~ 1.5 mW.

Holes were probed in fluorescence excitation with the same lasers used for burning, but the power was attenuated by a factor varying from 10^2 to 10^4. The fluorescence signal of the hole was detected with the PM. To separate the fluorescence signal from the scattered laser light, long-wavelength pass filters were used (Schott RG 665, total thickness ~ 1.5 cm) such that $\lambda_{\mathrm{det}} \geq 665$ nm.

The ‘effective’ homogeneous linewidth Γ'_{hom} was determined by extrapolating the holewidths, measured as a function of burning-fluence density Pt_{b}/A, to $Pt_{\mathrm{b}}/A \to 0$ [12,13]. The holewidths were well fitted with Lorentzian curves. The extrapolated value of the holewidth we call $\Gamma_{\mathrm{hole},0}$. Since in these experiments the burning time t_{b}, the delay time t_{d} and the probe time t_{p} were within the same order of magnitude, the expression for Γ'_{hom} reduces to [13]:

$$\Gamma'_{\mathrm{hom}}(t_{\mathrm{d}}) = \tfrac{1}{2}\Gamma_{\mathrm{hole},\,Pt/A \to 0}(t_{\mathrm{b}}, t_{\mathrm{d}}) - \Gamma_{\mathrm{laser}}\,. \qquad (1')$$

The bandwidth of the laser Γ_{laser} was either ~ 2 MHz (dye laser) or ~ 30 MHz (diode laser).

3. Results and discussion

3.1. Energy transfer within the $S_1 \leftarrow S_0$ 0–0 band

The absorption spectrum of H_2Ch in PS at 1.2 K is shown in Fig. 1a. The bands are inhomogeneously broadened with widths of ~ 200 cm^{-1}. The $S_1 \leftarrow S_0$ 0–0 band has its maximum at ~ 636 nm and is very intense compared to the vibronic bands. Its width, position and shape remain constant up to concentrations of 6×10^{-3} M, an indication that no aggregation occurs.

Broad-band fluorescence spectra of H_2Ch in PS at 1.5 K are displayed in Fig. 1b for three concentrations. The samples were excited in the Soret band, at $\lambda_{\mathrm{exc}} = 388$ nm and the spectra are normalized on the maximum of the $S_1 \to S_0$ 0–0 band. To prevent effects of re-absorption, only samples with low optical densities, OD ≤ 0.6, were used. At the lowest concentration, $c = 1 \times 10^{-5}$ M, the maximum of the 0–0 fluorescence band is at ~ 636 nm, equal to that of the 0–0 band in absorption, which indicates that there is no ET at this concentration and that the electron–phonon coupling strength is weak for H_2Ch in PS. At higher concentrations, the position of the 0–0 emission band shifts to the red and the band becomes narrower. Molecules excited in the blue

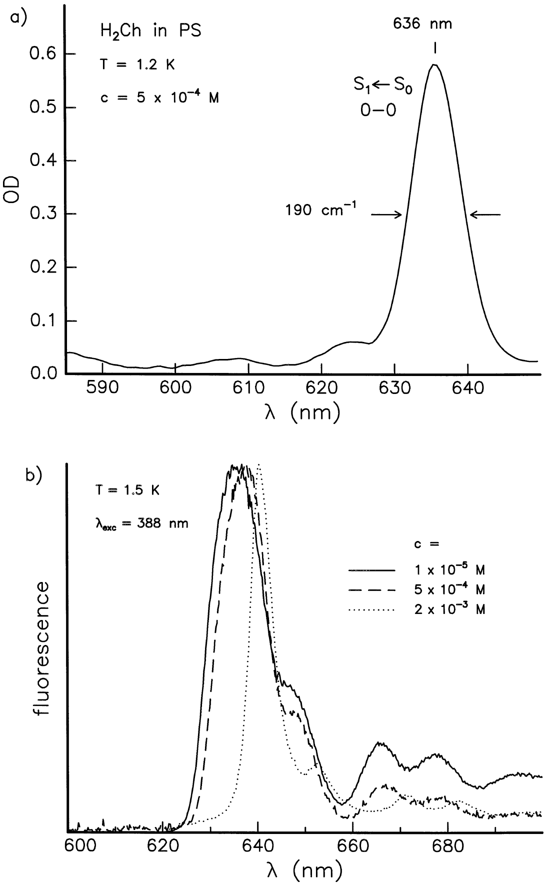

Fig. 1. (a) Absorption spectrum at 1.2 K of free-base chlorin (H_2Ch) in polystyrene (PS) at concentration $c = 5 \times 10^{-4}$ M. (b) Broad-band fluorescence spectra at 1.5 K of H_2Ch in PS, for three different concentrations. The red-shift and narrowing of the bands observed at higher concentrations is due to 'downhill' energy transfer.

part of the 0–0 band transfer their excitation energy to the red part through overlap of their phonon side-bands. Since ET competes with fluorescence, and the rate of ET increases with concentration, the fluorescence band shifts to the red and its width decreases with concentration. The spectra provide evidence that 'downhill' ET within the 0–0 band occurs for concentrations $c \geq 5 \times 10^{-4}$ M. This is supported by the observation of broad side-holes in the red wing of the 0–0 band formed after burning zero-phonon holes in the blue (not shown). These broad side-holes become more prominent with increasing concentration [23].

3.2. Spectral diffusion induced by 'downhill' energy transfer

3.2.1. Concentration- and excitation wavelength dependence of Γ'_{hom} at 1.2 K

To check to what extent 'downhill' ET is reflected in the 'effective' homogeneous linewidth $\Gamma'_{\rm hom}$, we have performed high-resolution HB experiments as a function of concentration and excitation wavelength with $t_{\rm d} = 300$ s. In Fig. 2 the results are shown for $\Gamma'_{\rm hom}$ as a function of concentration, for various excitation wavelengths. In the red wing of the 0–0 band, at 643 nm, $\Gamma'_{\rm hom}$ is constant and does not depend on concentration. Towards the blue, $\Gamma'_{\rm hom}$ increases linearly with concentration, the slope $\mathrm{d}\Gamma'_{\rm hom}/\mathrm{d}c$ becoming steeper at shorter $\lambda_{\rm exc}$. For $c \to 0$, all curves extrapolate to the same value of $\Gamma'_{\rm hom}$. We conclude that in the red wing of the 0–0 band no ET occurs at any concentration. At shorter wavelengths, and concentrations higher than $c = 1 \times 10^{-5}$ M, an effect caused by 'downhill' ET is observed. However, if the results were determined by direct ET of the Förster type [24,25], we would expect a quadratic dependence of $\Gamma'_{\rm hom}$ on concentration, instead of the linear dependence.

A second problem concerns the shape of the holes. In Fig. 2, three holes are depicted which were burnt at the corresponding excitation wavelengths in the most concentrated sample ($c = 6 \times 10^{-3}$ M).

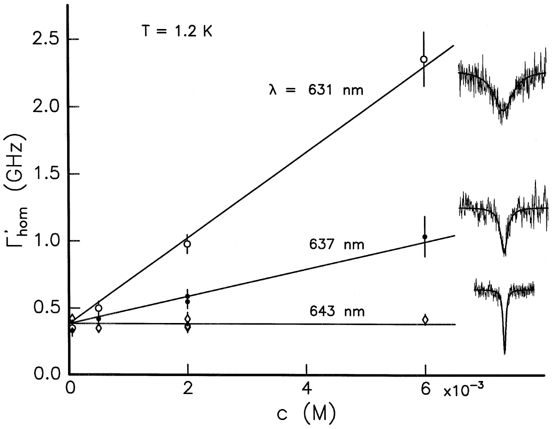

Fig. 2. 'Effective' homogeneous linewidth $\Gamma'_{\rm hom}$ as a function of concentration for H_2Ch in PS at $T = 1.2$ K and $t_{\rm d} \approx 300$ s, for three excitation wavelengths. Three holes are shown at their corresponding burning wavelengths, for $c = 6 \times 10^{-3}$ M; they have been fitted with Lorentzian curves.

The shape of these holes is well fitted with a Lorentzian curve, even in the extreme blue wing of the band, at the highest concentration. As shown by Förster [26], Inokuti and Hirayama [27], and Blumen and Silbey [28], the decay of the excited state due to dipolar Förster transfer is given by $\exp(-bct^{1/2})$ (where b is a constant and c is the concentration of acceptors). The overall decay is, therefore, given by $\exp(-t/\tau_{\mathrm{fl}})\exp(-bct^{1/2})$ which leads to a non-Lorentzian lineshape. However, for small c, the lineshape can be approximately fit by a Lorentzian whose width increases as c^2. For larger c, the line has a more sharply peaked shape and its FWHM is a complicated function of c.

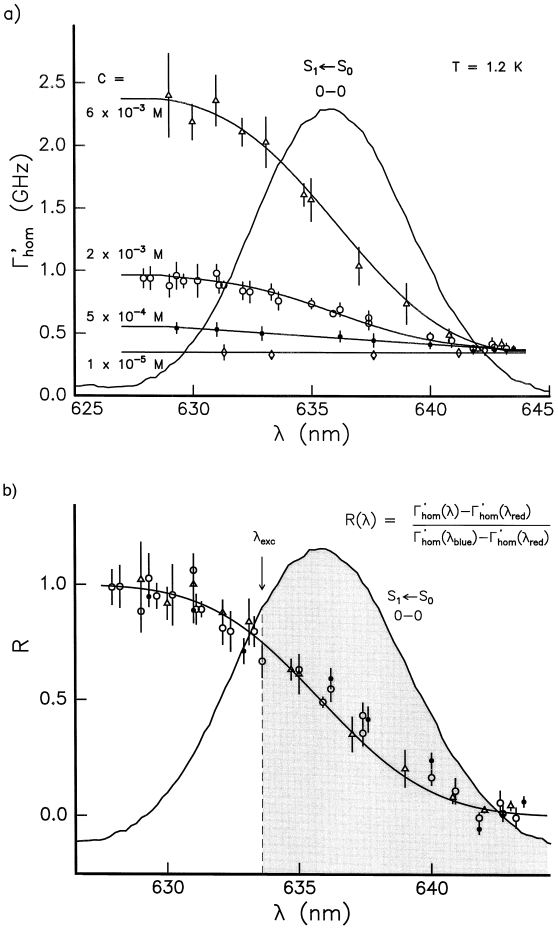

Fig. 3. (a) Excitation wavelength dependence of Γ'_{hom} for H_2Ch in PS at 1.2 K, for four concentrations. The $S_1 \leftarrow S_0$ 0–0 absorption band is also shown. The curves through the data are guide lines for the eye. (b) Ratio R as defined in the figure vs. λ. The data points are taken from (a). The $S_1 \leftarrow S_0$ 0–0 band is also shown. The curve traced through the data is not a fit, but represents the integral of the absorption band to the red of λ_{exc} (shaded area).

Fig. 3a shows the dependence of Γ'_{hom} on excitation wavelength λ_{exc} for four concentrations, superimposed on the $S_1 \leftarrow S_0$ 0–0 absorption band. At the lowest concentration, Γ'_{hom} has a small value and is independent of λ_{exc}. At higher concentrations, the data follow S-shaped curves which become more pronounced the higher the concentration. The S-shaped curves, which have been traced as guides for the eye, all coalesce in the red wing of the 0–0 band where they become horizontal. At the maximum of the 0–0 band, the slopes of the curves are steepest, while on the blue wing they become again horizontal. This behaviour indicates that we observe an effect caused by 'downhill' ET from molecules absorbing in the blue side of the absorption band to molecules absorbing towards the red.

We have normalized the curves of Fig. 3a in the following way. For each curve, a ratio R was calculated, defined as

$$R(\lambda) = \frac{\Gamma'_{\mathrm{hom}}(\lambda) - \Gamma'_{\mathrm{hom}}(\lambda_{\mathrm{red}})}{\Gamma'_{\mathrm{hom}}(\lambda_{\mathrm{blue}}) - \Gamma'_{\mathrm{hom}}(\lambda_{\mathrm{red}})}, \tag{2}$$

such that $R(\lambda_{\mathrm{red}}) = 0$ and $R(\lambda_{\mathrm{blue}}) = 1$, where λ_{red} is the longest- and λ_{blue} the shortest excitation wavelength used. The result is shown in Fig. 3b. All data now fall on a single curve because of the linear dependence of Γ'_{hom} on c (Fig. 2). The curve, which is not a fit to the data, is well approximated by an error function $F(\lambda_{\mathrm{exc}})$ representing the normalized area of the inhomogeneously broadened 0–0 band (assumed to have a Gaussian shape, $I_{\mathrm{inh}}(\lambda)$) at wavelengths longer than λ_{exc}:

$$F(\lambda_{\mathrm{exc}}) = \int_{\lambda_{\mathrm{exc}}}^{\lambda_{\mathrm{red}}} I_{\mathrm{inh}}(\lambda)\,\mathrm{d}\lambda. \tag{3}$$

Therefore, $cF(\lambda_{\mathrm{exc}})$ represents the number of molecules absorbing at wavelengths longer than λ_{exc}. Obviously, the results show that all these red-absorbing molecules are involved in the effect observed, and in such a way that

$$\Gamma'_{\mathrm{hom}} \propto cF(\lambda_{\mathrm{exc}}). \tag{4}$$

When trying to explain this result with Förster's ET mechanism we are confronted with two problems:

(1) If Γ'_{hom} were given by direct ET via the

overlap of the phonon-side bands of donor and acceptor molecules, we would expect Γ'_{hom} to be proportional to the number of acceptor molecules with transition frequencies between ~ 10 and 60 cm^{-1} (up to about twice the frequency of the phonon side-bands) to the red of λ_{exc} and, therefore, to follow approximately the inhomogeneous absorption band. This is not in accordance with Fig. 3.

(2) To be consistent with a reasonable Förster radius of 30–80 Å [29,30], we would expect the values of Γ'_{hom} in the blue wing of the band for the highest concentration to be only ~ 200 MHz larger than those for the lowest concentration, but the measured values are much larger. Thus, the major part of the effect is not directly caused by ET, but only *indirectly*.

To explain our results, we propose that Γ'_{hom} is predominantly determined by another mechanism: SD induced by ET, in addition to 'normal' SD. The idea is that during the excited-state lifetime $\tau_{\mathrm{fl}} \approx 8$ ns of H_2Ch, excitation energy is transferred stepwise from the excited probe molecules to others absorbing further to the red. At each step, part of the mismatch of the excitation energy of donor and acceptor molecules ΔE will be released as 'heat' into the glass. The number of transfer steps and the available energy will increase with the concentration of H_2Ch molecules absorbing at energies lower than the initial excitation wavelength (i.e., to the red of it). By this mechanism, a large part of the glass in the neighbourhood of H_2Ch molecules is explored. If during this exploration, the excitation finds itself close to one or more TLSs, energy can be stored into them. The activated TLSs will subsequently relax and give rise to 'extra' SD, after ET has taken place, i.e. over many orders of magnitude in time. As a consequence, we would expect Γ'_{hom} to increase with delay time, even at $T \to 0$. This increase of Γ'_{hom} with delay time is expected to be larger for higher concentrations and towards the blue within the 0–0 absorption band.

3.2.2. Increase of Γ'_0 with delay time and concentration

To verify our prediction, we have measured Γ'_{hom} vs. temperature, for two concentrations ($c = 1 \times 10^{-5}$ and 2×10^{-3} M) and two delay times ($t_{\mathrm{d}} = 100$ μs and 300 s), in the blue wing ($\lambda_{\mathrm{exc}} = 633$ nm) of the 0–0 band. The results are shown in Fig. 4. For each concentration and delay time, the data follow a power law of the form of Eq. (1), characteristic for doped organic glasses. At the lowest concentration, the two curves for the two delay times extrapolate to a common residual linewidth $\Gamma_0 = (2\pi\tau_{\mathrm{fl}})^{-1} \approx 20$ MHz, for $T \to 0$, determined by the fluorescence lifetime $\tau_{\mathrm{fl}} \approx 8$ ns of H_2Ch [21]. The coupling constant $\boldsymbol{a} = a_{\mathrm{PD}} + a_{\mathrm{SD}}(t_{\mathrm{d}})$ increases with delay time and represents PD and 'normal' SD [12,13,31]. At the higher concentration, however, the two curves extrapolate to $\Gamma'_0 > \Gamma_0$, the value of Γ'_0 being larger for the longer delay time. Thus, these results provide evidence that through 'heat' released into the TLSs of the glass, Γ'_{hom} is predominantly determined by 'extra' SD indirectly induced by ET.

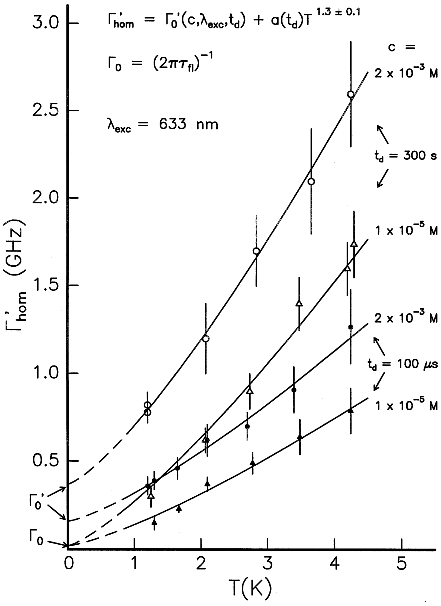

Fig. 4. Temperature dependence of Γ'_{hom} for H_2Ch in PS at the blue wing of the 0–0 band, for two concentrations and delay times. All curves follow a $T^{1.3}$ dependence. At $c = 1 \times 10^{-5}$ M, the two curves extrapolate to a common value $\Gamma_0 = (2\pi\tau_{\mathrm{fl}}) \approx 20$ MHz, with $\tau_{\mathrm{fl}} \approx 8$ ns the fluorescence lifetime of H_2Ch. At $c = 2 \times 10^{-3}$ M, the curves extrapolate to $\Gamma'_0 > \Gamma_0$, with Γ'_0 increasing with delay time.

4. Conclusions

The red-shift and narrowing of the 0–0 band of the fluorescence spectrum of H_2Ch in PS with increasing concentration of the probe molecules show that ‘downhill’ ET occurs in this system for $c \geq 5 \times 10^{-4}$ M.

The shape of the narrow holes burnt into the 0–0 band, the values of Γ'_{hom}, and the dependence of Γ'_{hom} on concentration and excitation wavelength at 1.2 K prove that Γ'_{hom} is not primarily determined by direct ET of the Förster type, but mainly by slow SD triggered by fast ‘downhill’ ET processes. We summarize here the experimental findings that led us to propose this mechanism:

(1) Γ'_{hom} depends linearly on concentration, and not quadratically.

(2) Holes have Lorentzian shapes, independent of concentration and excitation wavelength.

(3) Γ'_{hom} increases towards the blue of the 0–0 band following an S-shaped curve that indicates that all molecules absorbing to the red of λ_{exc} participate in the broadening process.

(4) The values of Γ'_{hom} are much larger than those expected from a Förster radius of 30–80 Å.

Concluding that ET → SD is the process responsible for these observations, we expected the value of Γ'_0 to increase with delay time at concentrations for which ET takes place. This has indeed been confirmed.

A further quantitative analysis of the separate contributions of τ_{fl}, ET, and ET → SD to the residual linewidth, and of PD, ‘normal’ SD, and ET → SD to the coupling constant in the expression of Γ'_{hom} will be presented in a subsequent paper [31].

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