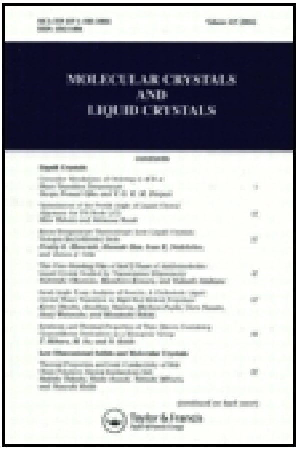

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Interacting Tunneling States: A Hole-Burning Study of Spectral Diffusion

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INTERACTING TUNNELING STATES: A HOLE–BURNING STUDY OF SPECTRAL DIFFUSION

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Abstract Performing long–time thermal equilibrium measurements of spectral diffusion broadening of photochemical holes, we recently established an algebraic time dependence of the dynamics of two–level system (TLS), which we interpret in terms of TLS–TLS coupling (Phys. Rev. Lett. **76** (1996) 2085). For this purpose we utilized the continuous cooling property of our ^{3}He/^{4}He cryostat. To extend the accessible temperature range we have constructed a cryostat which allows optical experiments lasting several weeks at temperatures of 1.5 K and above. Here we report data obtained at 2.0 K in a time range from 10 s to 10^6 s. Interpreted in the framework mentioned above, the results indicate the onset of a thermally induced break–up of the TLS–TLS coupling.

INTRODUCTION

Since the first observation of persistent spectral holes in organic glasses [1] spectral hole–burning has been extensively used for the investigation of the low temperature properties of amorphous solids (see for example reviews [2, 3] and references therein). The phenomenon of spectral diffusion, which describes the time and temperature dependence of spectral line widths in amorphous hosts, makes optical hole–burning a very efficient tool for the investigation of the dynamics of tunneling states in glasses also referred as two–level systems (TLS)[4] (see also reviews [2, 3]). This method is capable of yielding basically the same information as the more traditional approaches of measuring the time dependent specific heat or heat release [5]. For the study of very slow TLS dynamics, however, the hole–burning approach bears principal advantages: Both of the mentioned calorimetric methods are based on detecting the decay of a thermal non–equilibrium situation [6]. They are therefore limited to a time range where this decay is still strong enough to be detected (usually by measuring temperature changes). For the observation of the diffusional broadening of spectral holes, however, it is not necessary to disturb thermal equilibrium. Furthermore suitable probe molecules allow the creation of persistent holes, so that the experimental time range is virtually unlimited.

We have recently shown that the measurement of spectral diffusion in thermal equillibrium and at very long times, i. e. up to 10^6 s, is capable of providing new insight into the mutual interactions of TLS degrees of freedom and the resulting dynamics [7]. In order to suppress contributions from thermal relaxation, the sample has to be kept at constant low temperature for several weeks before hole–burning and starting the measurements [5, 7]. Therefore we have constructed a cryostat that allows optical experiments under continuous cooling conditions in the temperature range between 1.5 K and 4.2 K for several weeks:

EXPERIMENTAL

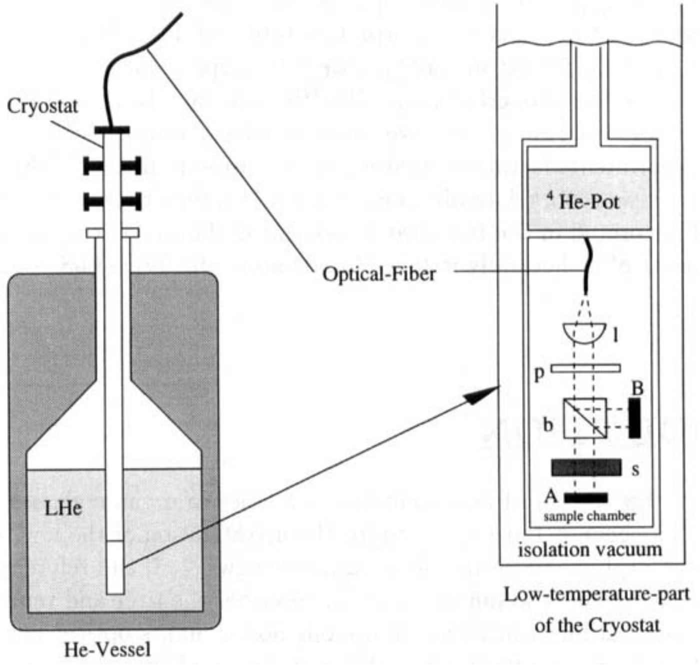

Figure 1: The dipstick cryostat inside a He-vessel and the optical path in its low temperature part

We use a conventional dipstick cryostat [9] which is constructed to perform measurements in a normal He storage vessel. The He consumption of the cryostat is less than 2 l LHe per day, which yields in the case of a 100 l He-vessel an experimental time of almost 50 days. A separated sample chamber mounted on the cooling plate of the He-pot filled with a few mbar of He gas guarantees good thermal coupling of the sample. This also enables temperature measurement via a vapor pressure thermometer. The temperature regulation is performed with an electronic circuit between a carbon resistor mounted next to the sample and an ohmic heater on top of the ^{4}He-pot. This way we achieved a temperature stability of better than ± 0.01K during the whole experiment.

Holes are burned and read with a single frequency cw dye laser (Coherent CR-699-29 Autoscan) whose output is fed into the sample chamber with a single-mode fiber. The end of the fiber is positioned in the focus of a 10mm lens (l) which collimates the beam to a diameter of 2.5mm. The transmitted light is detected with a photodiode (A) (Siemens BPW-33) directly inside the sample chamber. The idea of detecting the optical signal with photodiodes at low temperature was previously published in [10].

An advantage of this arrangement is the improvement of the detection limit of the photodiodes with temperature due to the lower thermal noise. The noise in the whole detection circuit is given by shot noise generated in the photodiode and thermal noise generated in the leads to the current amplifiers. The shot noise consists of contributions from the photocurrent itself and the leakage current due to the applied bias voltage. Shot noise, however can be neglected in comparison to the thermal noise. The NEP for the whole detection circuit is estimated to be ≤ 1 pW.

The polarizer (p) behind the lens (l) ensures that the polarization of the light remains constant during burning and all following reading processes. In combination with the slowly rotating polarization of the light caused by the fiber this gives rise to variations in light power. To correct these variations, the transmission signal is normalized with the incident light power by dividing the signal of diode A by the signal of diode B. The Power monitoring diode B is mounted next to a beam splitter (b) located between polarizer and sample (s).

In the experiment at 2 K the holes were burnt with about 0.1 μW laser power which was reduced by a factor of 2000 for reading the spectra. At this power level no light-induced modification of the hole profile was observed after 50 reading cycles.

DISCUSSION

In ref. [7] we reported a much stronger than logarithmic increase of the diffusional hole broadening at times exceeding hours. These data were obtained for the polymer glass PMMA doped with the organic dye phthalocyanine (H_2PC) at temperatures of 0.5 K and 1.0 K, respectively. We are extending the temperature range of our experiments which is necessary for a reliable interpretation of the observed algebraic time dependence. Here we report data on the same amorphous host measured at 2.0 K using the molecule tetra-phenyl-porphin (TPP) as an optical probe for TLS dynamics. Since time resolved diffusional hole broadening scales approximately linear with temperature [5], fig.2 shows the three datasets in a temperature normalized plot to focus on the curve shapes rather than the absolute values of the broadening. The figure clearly shows that the new (2 K) data looks qualitatively similar to the two 'low-temperature' datasets. The main feature of 2.0 K data is the same as in 0.5-1 K data – they are clearly nonlogarithmic and, as follows from the quantitative analysis,

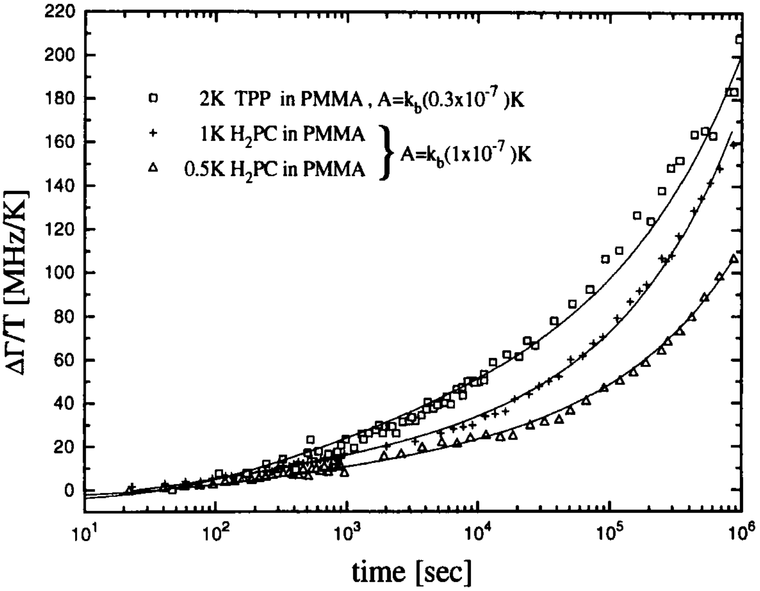

Figure 2: *Observed hole-broadening at 0.5 K, 1 K and 2 K. The different curve shapes are obvious and can be interpreted as a thermal induced break up of the TLS–TLS coupling in the framework of our model. For details of the fitparameter A, see text.*

obey a $\sqrt{t}$ law at long times. But the 2.0 K data do not follow precisly the trend that could be expected from the lower temperature data: Instead of further increase with increasing temperature, the long-time 'nonlogarithmic' slope of the 2.0 K data displays the opposite tendency. From this observation it is obvious that the temperature dependence of spectral diffusion cannot be accounted for by a straightforward scaling of TLS relaxations with temperature and the TLS density of states does not remain constant with changing temperatures above 2.0 K.

In ref. [7] we proposed an ansatz for the TLS density of states, which is motivated by a scenario of strongly interacting TLS [8]:

$$P_C(\Delta, \Delta_0) d\Delta d\Delta_0 = \bar{P} \cdot \left[\frac{1}{\Delta_0} + \frac{A}{\Delta_0^2}\right] d\Delta d\Delta_0, \tag{1}$$

Here Δ is the TLS energy asymmetry while Δ_0 represents the tunneling matrix element. The Δ_0^{-1} term corresponds to the standard tunneling model, which results in a logarithmic time dependence of spectral diffusion. The second term accounts for the dynamics of coupled TLS. Its exponent has been chosen to agree with theoretical predictions about TLS–TLS interactions (refs.[11, 12, 13]). It results in a $\sqrt{t}$ evolution of the hole broadening. The coeffizient A is a 'weightfactor' of the two terms; it determines the fraction of interacting TLS within the whole ensemble and serves as free fit parameter in our treatment. Parameter A has energy dimension and, sine the square of the off-diagonal matrix element Δ_0 constitutes a transition propability, it determines the range where a crossover from a logarithmic ($1/\Delta_0 \gg A/\Delta_0^2$) to an algebraic ($1/\Delta_0 \ll A/\Delta_0^2$) time dependency occurs.

The fit of 0.5 K and 1.0 K data with use of the TLS distribution function (eq. 1) results in a single consistent set of parameters within 10 %, with $A = k_b(1.0 \pm 0.1) \cdot 10^{-7})$ K, i.e. the distribution function remains constant for both temperatures. At 2.0 K, however, the situation changes and agreement between model curve and experimental data can be achieved by decreasing A to a value of $k_b \cdot (0.3 \cdot 10^{-7})$ K. As fig. 2 shows, this results in a good description of the data. Within a model of coupled TLS we have to assume an interaction energy ϵ, that prevents a single TLS from flipping independently. For the TLS sub-ensemble with $\epsilon \ll k_B T$ this "locking" results in coherent $\sqrt{t}$ dynamics according to the theoretical predictions of [11, 12, 13]. With increasing temperature, however, this coherence has to break up for an increasingly larger fraction of the TLS ensemble. Within this framework we interpret the decrease of the "weight parameter" A with the assumption that at 2.0 K a thermal energy level is reached where the loss of coherence becomes substantial enough to distinctly modify the time evolution of spectral diffusion. Since the TLS-TLS interaction energy ϵ itself does not enter our empirical model, our analysis only yields a decrease of the fraction of "locked" TLS given by A/Δ_0^2.

In summary we have extended the temperature range accessible to our long-term spectral diffusion experiments. The result obtained at 2.0 K distinctly deviates from the behavior observed at $T \geq 1.0$ K. Within our empirical model description we can interpret this modified diffusional hole broadening by assuming a thermally induced break-up of the coherent dynamics of coupled TLS.

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