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Spectral diffusion decay of spontaneous echoes in disordered systems

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Résumé. — La relaxation des échos spontanés dans un système de spin $\frac{1}{2}$ ou de systèmes à 2 niveaux en interaction est traitée dans le cadre du modèle Klauder-Anderson. L'hypothèse d'une distribution hyperbolique des temps de relaxation T_1 — ou « bruit en $1/f$ » — conduit à une loi de relaxation très générale en $\exp(-\gamma T\tau)$ où T est la température, τ l'intervalle entre les deux impulsions et γ un coefficient ne dépendant que de l'interaction moyenne entre systèmes à 2 niveaux excités thermiquement.

Abstract. — In a system of two level systems or $\frac{1}{2}$ spins, the relaxation of spontaneous echoes is obtained in the framework of the Klauder-Anderson scheme. The assumption of a hyperbolic distribution relaxation times T_1 (or « $1/f$ noise ») leads to a general relaxation law : $\exp(-\gamma T\tau)$ where T is the temperature, τ the delay time between the two pulses and γ a coefficient depending only on the average interaction between the thermally excited two level systems.

Introduction. — It is now well established that in pure glasses low energy excitations exist which are termed tunnelling modes. These tunnelling modes are described as two level systems exhibiting an almost constant density of states for splitting energies below 1 K. They are also strongly coupled to the phonon field and possess a very broad distribution of their T_1 . In addition to these two level defects, OH doped glasses introduce electric dipole moments with more or less the same characteristics as the two level systems that is a uniform density of states as well as broad distribution of their T_1 [1, 2]. Finally, these defects interact among themselves *via* the phonon field rather than *via* the electric field [2]. Therefore in pure as well as in doped glasses a set of two level defects exists which are coupled by interactions varying as r^{-3} : The dynamics of these two level systems may be treated by the same methods used for systems of true spins $S = 1/2$ coupled by dipolar magnetic interactions. Following the scheme used by Klauder-Anderson [3] for spectral diffusion in spin systems, we divide the two level systems into two classes A and B : A contains the 2 level defects whose splitting energy is in resonance with the applied field-acoustic (resp. electric) for the acoustic (resp. electric) dipolar echoes. The number of A defects is small and largely diluted in the sample and the interactions between them may be ignored. But they are coupled [4] to

the B defects *via* the elastic strain of the medium usually described by a hamiltonian $J_{ij} S_z^i S_z^j$ where the S_z^i are the diagonal Pauli operators in the representation of the two states of the defects. Transitions between the two levels of the B defects induce fluctuating local fields at the level of the A defects. The frequency distribution of the excited packet of A defects therefore becomes broader due to the dephasing effect of fluctuations of the local field. It is assumed that the system is of T_1 -type : i.e. the rate of fluctuations is caused by the spin-phonon coupling. This scheme constitutes the basic model proposed first by Black and Halperin [5] for explaining the phonon echoes first discovered by Golding and Graebner [6]. However, they focus attention on the short time regime where τ , the delay time between the two pulses, is shorter than the shortest thermal B spin T_1 of the T_1 -distribution : In this regime they found that the decay of the echo amplitude is given by :

$$E(2\tau) = \exp(-\alpha T^4 \tau^2) \quad (1)$$

where α is a coefficient taking into account all the ingredients of the coupling between the 2 level defects. This regime has been studied in previous experiments by Golding and Graebner [6] and Schickfus *et al.* [7] but several recent experiments of Bernard *et al.* [1, 8] at lower temperatures of dielectric echoes in glasses,

and in smoky quartz [9], indicate strongly that the decay function $E(2\tau)$ varies as

$$E(2\tau) = \exp(-\gamma T\tau). \quad (2)$$

The purpose of this article is to derive in a simple way this type of variation. This law emerges in the regime where τ is *inside* a broad *hyperbolic* distribution $1/T_1$ of the defects T_1 . In this regime remarkably few parameters enter into the coefficient γ : it is simply the product of the spectral and volume density of B spins N_B (in $\text{K}^{-1} \text{m}^{-3}$) and the average local dipolar field on spin A. Nothing refers to the detailed mechanism of relaxation as the one phonon assisted tunnelling motion. Neither the cut-off in the hyperbolic distribution $1/T_1$ nor the details of the one-phonon process are important. We believe that this regime is much more common at very low temperature than the one described by (1), particularly if one considers the hyperbolic distribution as the $1/f$ noise which is often invoked at low temperature (Clarke and Voss [10]).

In the Black and Halperin paper [5], this regime is qualified as « a roughly exponential decay bounded from below by $\exp(-\pi^{-1/3} m T_1 \tau)$ » in their notation. However the expression and its derivation are not satisfactory because while they use the cut-off for the T_1 -distribution as well as an extrapolation of the coefficient m from the short time regime to this intermediate regime [11].

1. Echo amplitude. — Let us derive the expression for the spontaneous echo amplitude $E(2\tau)$ of a collection of two level systems characterized by an energy splitting E and a relaxation rate $W = 1/T_1$, with a density of states per unit energy and volume $N(E)$ and a joint distribution $p(W, E)$.

The two pulse echo amplitude is given by Klauder and Anderson [3] and Mims [12]:

$$E(2\tau) = \left\langle \left\langle \exp \left\{ i \int_0^{2\tau} S(t) \Delta\omega_i(t) dt \right\} \right\rangle_i \right\rangle_j \quad (3)$$

where

$$S(t) = \begin{cases} 1 & \text{if } 0 < t < \tau \\ -1 & \text{if } \tau < t < 2\tau \end{cases}$$

and $\hbar \Delta\omega_i(t) = \sum_{j \in B} J_{ij} S_z^j(t)$ is the time fluctuating local field on the i -th spin A. As S_z^j is the standard Pauli matrix, we write:

$$J_{ij} = \frac{C_{ij} \mu_i \mu_j}{R_{ij}^3} \quad (4)$$

where μ_i are the dipole moments and C_{ij} the coupling tensor.

$\langle \dots \rangle_j$ represents the spatial average over B defects and $\langle \dots \rangle_i$ the spatial average over A spins, is replaced by a time average over all histories of B spins. Since the two level systems possess two parameters E and W , there are additional averages to be considered here.

The time average is performed through an uncorrelated sudden-jump model where the *excitation* flipping rate W_1 can be different from the *desexcitation* flipping rate W_2 . In thermal equilibrium

$$W_1/W_2 = \exp(-E/k_B T)$$

and $W = W_1 + W_2$.

This average as well as the spatial average of B spins has been calculated by Hu and Walker [13] using the $1/r^3$ range of the interaction energy and the details of the calculation will not be repeated here. Let us write:

$$E(2\tau) = \exp \left\{ -\frac{\pi}{6} C_{\text{rms}} \mu_A f(2\tau) \right\} \quad (5)$$

where $C_{\text{rms}} = \sqrt{\langle C_{ij}^2 \rangle}$ is an average value of the coupling constant and μ_A another average dipole moment of A defects. Both factors define an average dipolar local field.

The Laplace transform of $f(2\tau)$, $F(\sigma)$ has been derived in reference [13] for E and W fixed (see (15) and (16) in this reference) and has the following expression

$$[4 W_1 W_2 / (W_1 + W_2)] [(\sigma + 2 W_1 + 2 W_2) (\sigma + 2 W_1) (\sigma + 2 W_2) \sigma^3]^{-1/2}. \quad (6)$$

By considering the additional distribution of E and W : $p(E, W)$ as well as the detailed balance of W_1 and W_2 we directly obtain:

$$F(\sigma) = \iint p(E, W) dE dW \cdot \mu_B \text{sech}^2(E/2kT) \times \\ \times 2 W [(\sigma + 2 W) (\sigma + 2 W / (1 + e^{-E/kT})) (\sigma + 2 W / (1 + e^{E/kT})) \sigma^3]^{-1/2} \quad (7)$$

2. The hyperbolic noise. — Suppose now that the B-two level defects have a hyperbolic distribution law for their relaxation time T_1 or in an equivalent way for their relaxation frequency $W = 1/T_1$:

$$p(W, E) = \begin{cases} n(E) \frac{1}{W} & \text{if } W_{\text{MIN}} \leq W \leq W_{\text{MAX}} \\ 0 & \text{elsewhere} \end{cases} \quad (8)$$

and a constant spectral density $n(E) = n_0$ for $E \leq E_{\text{MAX}}$.

The argument of the decay function $F(\sigma)$ has now a very simple form in the dimensionless variables $x = E/kT$ and $u = 2W/\sigma$:

$$F(\sigma) = \frac{n_0 k_B T}{\sigma^2} \mu_B \int_0^{E_{\text{MAX}}/T} dx \operatorname{sech}^2(x/2) \times \int_{W_{\text{MIN}}/\sigma}^{W_{\text{MAX}}/\sigma} du [(1+u)(1+u/(1+e^{-x}))(1+u/(1+e^x))]^{-1/2}. \quad (9)$$

At very low temperatures, $T \ll E_{\text{MAX}}$ and the $\operatorname{sech}^2(x/2)$ enables x to be integrated to infinity. 3 different regimes now appear, defined by the value of σ compared to the cut-off W_{MAX} and W_{MIN} :

2.1 SHORT TIME-LARGE σ ($\sigma \gg W_{\text{MAX}}$). — u is always less than one since $\sigma \gg W_{\text{MAX}}$. The second integral gives W_{MAX}/σ and $F(\sigma)$ varies as $T/\sigma^3 W_{\text{MAX}}$. The inverse Laplace transform $\ln E(2\tau)$ varies as $-\tau^2 TW_{\text{MAX}}$. For the one-phonon assisted tunnelling

relaxation $W_{\text{MAX}} \approx T^3$ which leads to the $\tau^2 T^4$ law obtained in reference [5].

2.2 LONG TIME-SMALL σ ($\sigma \ll W_{\text{MIN}}$). — $u \gg 1$ and the u -integral can be approximated by $\sigma^{1/2} \operatorname{ch} x/2$ which gives a $F(\sigma)$ varying as $\sigma^{-3/2}$. The inverse Laplace transform gives the usual *central limit* regime $\ln E(2\tau)$ as $-\tau^{1/2}$ (Ref. [11]).

2.3 INTERMEDIATE REGIME : $W_{\text{MIN}} \ll \sigma \ll W_{\text{MAX}}$. — The convergence of the u -integral is possible when $W_{\text{MIN}}/\sigma = 0$ and $W_{\text{MAX}}/\sigma = \infty$. Using

$$\int_0^\infty \frac{dx}{\operatorname{ch}^2 \frac{x}{2}} \int_0^\infty du \left[(1+u) \left(1 + \frac{u}{1+e^{-x}} \right) \left(1 + \frac{u}{1+e^x} \right) \right]^{1/2} = 2.635 \quad (10)$$

we get for $F(\sigma)$:

$$F(\sigma) = 2.635 \frac{n_0 k_B T}{\sigma^2} \mu_B. \quad (11)$$

Now the inverse Laplace transform gives for the echo amplitude :

$$E(2\tau) = \exp \left\{ -2.635 \frac{C_{\text{rms}} \mu_A \mu_B n_0 kT}{h} \tau \right\}. \quad (12)$$

As $n_0 kT$ can be interpreted as the average number of excited two level systems at temperature T , the product $C_{\text{rms}} \mu_A \mu_B n_0 kT$ represents the average interaction energy among the *thermally excited* two level systems : it is the inhomogeneous thermal linewidth of the problem. Very striking is the disappearance of any mechanism of relaxation process in (12) : this comes formally from the fact that τ is well inside the hyperbolic distribution law of relaxation times.

3. **Disordered substances.** — The original model of Anderson, Halperin, Varma and Philips [14] introduced two tunnelling parameters : Δ the asymmetry energy and the tunnelling coupling energy given by $\Delta_0 = \hbar\omega_0 e^{-\lambda}$. The energy splitting is obtained by the relation $E = (\Delta^2 + \Delta_0^2)^{1/2}$. It is assumed that Δ and λ are uniformly distributed between two limits

$\lambda_{\text{MIN}} < \lambda < \lambda_{\text{MAX}}$. The defect-lattice relaxation time $T_1(E, \lambda)$ is given in reference [5]

$$W = T_1^{-1}(E, \lambda) = AE^3 e^{-2(\lambda - \lambda_{\text{MIN}})} \coth(E/2 k_B T). \quad (13)$$

The distribution can be rewritten in terms of the variables E and W :

$$n(E, W) = \frac{\bar{n}}{2} \frac{1}{W} (1 - W/W_{\text{MAX}})^{-1/2}; \quad (14)$$

$$W_{\text{MIN}} < W < W_{\text{MAX}}$$

where W_{MAX} is obtained from (13) with $\lambda = \lambda_{\text{MIN}}$.

In addition of this distribution law the permanent elastic dipole moment μ_B is multiplied by the factor $\Delta/E = (1 - W/W_{\text{MAX}})^{1/2}$. The factor $(1 - W/W_{\text{MAX}})^{1/2}$ cancels in the product of the two expressions giving the hyperbolic distribution law (8) from which the exponential $\exp(-\gamma\tau)$ regime is derived. These are basically the standard assumptions necessary for analysing the echo measurements on doped glasses. From the expression (12) recast in the standard formula : $E(2\tau) = \exp(-2\tau/T_2)$, the phase memory decay time is defined :

$$T_2 = h/(1.31 C_{\text{rms}} \mu_A \mu_B kT) \quad (15)$$

which is interpreted as the inverse average interaction frequency among the populations of the thermally excited states of two level systems. Only the order of magnitude of these parameters are known : $C_{rms} \mu_A \mu_B = 10^{-35}$ ergs cm³ corresponding to an average elastic coupling energy $M = 1.5$ eV/defect (see Ref. [5]);

$$n_0 = 10^{32} \text{ ergs}^{-1} \text{ cm}^{-3} \quad \text{or} \quad 1.6 \times 10^{20} \text{ eV}^{-1} \text{ cm}^{-3}$$

from reference [15].

The product $C_{rms} \mu_A \mu_B n_0$ is dimensionless and is estimated to be 10^{-3} . From these values we get $T_2 = 10^{-5}$ s at $T = 10^{-2}$ K instead of 10^{-4} s at $T = 10^{-2}$ K from figure 4 of reference [1]. Taking into account the uncertainty of various parameters we conclude that the order of magnitude of T_2 from electric dipolar echoes is roughly described by the expression (15).

Since T_2^{-1} is linear in temperature, the lower curve of figure 4 of reference [1] is therefore well explained. As the limits of the hyperbolic distribution of T_1 do not enter in (15), they cannot be determined. However the limit of validity of this regime is reached at lower temperatures [17] when W_{MAX} is comparable to τ^{-1} .

One can consider crystalline materials doped with random dipolar impurities such as $\text{KCl} : \text{OH}$ as a similar situation to glasses but with clustering effect of dipoles at very low temperature. This case of

weak tunnelling is characterized by a distribution of tunnelling energies Δ_0 cut-off at some low energy Δ_c instead of ranging up to kT ($\Delta_c \ll kT$). This smaller value of the cut-off would reflect the collective nature of the tunnelling motion of clusters of dipoles. For these defects the noise distribution is purely hyperbolic since $W \ll W_{MAX}$ and the correction to the permanent elastic dipole is also $\Delta/E \simeq 1 + 0(W/W_{MAX})$ in such a way that the hyperbolic distribution is still valid. This type of weak tunnelling has been used in the analysis of the dielectric properties at very low temperature of ionic glasses BK7 [16].

4. Conclusion. — In summary, the regime of exponential decay which has been established here is based upon the assumption of a hyperbolic distribution of the relaxation time T_1 in addition to the standard ingredients of the Klauder-Anderson model namely long range r^{-3} interaction, and Poissonian noise. We emphasize the generality of this regime already observed for various disordered substances at very low temperature. The hyperbolic distribution of T_1 is usually attributed to the broad distribution of potential barriers for tunnelling motion. However since the spectral power of this noise is $1/f$ (f = frequency) the origin of this hyperbolic noise is perhaps more common than usually invoked in the context of disordered systems. Therefore the spontaneous echo decay measurement could therefore be an useful method for the investigation of this type of noise.

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