HETEROGENEOUS SPIN LATTICE RELAXATION IN DISORDERED SYSTEMS

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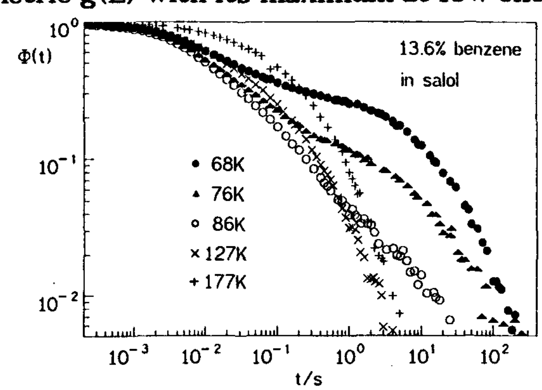
Disordered systems, e.g. organic glasses, are characterized by topological disorder. A distribution of intermolecular distances and orientations governs the local potentials. Subensembles of nuclear spins are coupled to different lattices. Generally, this results in a heterogeneous, i.e. non-exponential, spin lattice relaxation which can be described by a convolution of an exponential function with a distribution function $f(T_1)$ characterizing the disordered system. However, in the case of ^1H NMR fast spin diffusion averages the heterogeneous spin-lattice relaxation and in many cases an exponential decay, homogeneous relaxation, is found. This drawback is not given for ^2H NMR. Because of the smaller dipolar coupling among the deuterons spin diffusion is small. Thus, ^2H NMR is suited to detect heterogeneous relaxation; particularly, when deuterated mobile guest molecules are doped into a disordered matrix motional heterogeneities are revealed. Correlation time τ and activation energy E for the reorientation of the probe molecules are strongly dependent on the local environment. Assuming a thermally activated process a distribution g(E) results in a temperature dependent distribution $p(\tau)$ or $G(\ln\tau)$ which becomes broader when the temperature decreases.

By applying the standard relaxation theory (BPP) the heterogeneous spin lattice is given by two different Laplace transformations for the limiting cases $\omega\tau \ll 1$ and $\omega\tau \gg 1$, respectively. This is a consequence of the symmetry of the BPP formula with respect to $\ln\tau$. For $\omega\tau \ll 1$ the spin lattice relaxation is given by the Laplace transformation of the distribution of correlation times $p(\tau)$ and for $\omega\tau \gg 1$ by a Laplace transformation of the rate distribution P(r), where $r=1/\tau$.

For the condition $\omega\tau \sim 1$ a complex distribution $f(T_1)$ with at least two maxima determines the relaxation behaviour. Any asymmetric distribution function for $G(\ln\tau)$ results in different non-exponential relaxation for the two limiting cases. In contrast to other relaxation methods this sensitivity to a break of symmetry makes ^2H NMR studies particularly suited to detect details of a given distribution function and the deconvolution procedure is simplified by studying a wide temperature range. The method can be used to characterize disordered media like high and low molecular glasses, chaircoals or even proteins.

In the case of a homogeneous relaxation no symmetric relaxation minimum and no temperature independent slope $d\ln T_1/d(1/T)$ is given for the limiting cases.

^2H NMR spin lattice relaxation measurements covering a temperature range of 50-180K are shown. At lowest temperatures the spin lattice relaxation is studied over six decades in time indicating a broad distribution of relaxation times corresponding to an asymmetric g(E) with its maximum at low energies.



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