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Title:Distance measurements across randomly distributed nitroxide probes from the temperature dependence of the electron spin phase memory time at 240 GHz
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Abstract:At 8.5 T, the polarization of an ensemble of electron spins is essentially 100% at 2 K, and decreases to 30% at 20 K. The strong temperature dependence of the electron spin polarization between 2 and 20 K leads to the phenomenon of spin bath quenching: temporal fluctuations of the dipolar magnetic fields associated with the energy-conserving spin "flip-flop" process are quenched as the temperature of the spin bath is lowered to the point of nearly complete spin polarization. This work uses pulsed electron paramagnetic resonance (EPR) at 240 GHz to investigate the effects of spin bath quenching on the phase memory times (T<inf>M</inf>) of randomly-distributed ensembles of nitroxide molecules below 20 K at 8.5 T. For a given electron spin concentration, a characteristic, dipolar flip-flop rate (W) is extracted by fitting the temperature dependence of T<inf>M</inf> to a simple model of decoherence driven by the spin flip-flop process. In frozen solutions of 4-Amino-TEMPO, a stable nitroxide radical in a deuterated water-glass, a calibration is used to quantify average spin-spin distances as large as r¯=6.6nm from the dipolar flip-flop rate. For longer distances, nuclear spin fluctuations, which are not frozen out, begin to dominate over the electron spin flip-flop processes, placing an effective ceiling on this method for nitroxide molecules. For a bulk solution with a three-dimensional distribution of nitroxide molecules at concentration n, we find W∝n∝1/ r¯³, which is consistent with magnetic dipolar spin interactions. Alternatively, we observe W∝n3² for nitroxides tethered to a quasi two-dimensional surface of large (O ∼ 200 nm), unilamellar, lipid vesicles, demonstrating

that the quantification of spin bath quenching can also be used to discern the geometry of molecular assembly or organization. © 2012 Elsevier Inc. All rights reserved.

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Controlled terms:Deuterium - Distance measurement - Electron spin resonance spectroscopy - Electrons - Flip flop circuits - Magnetic moments - Magnetic resonance - Molecules - Paramagnetism - Quenching - Spin fluctuations - Spin glass - Spin polarization - Temperature distribution - Two dimensional
Uncontrolled terms:Bulk solutions - Decoherence - Dipolar magnetic fields - Electron spin polarization - Electron spins - Energy-conserving - Frozen solutions - High field - Lipid vesicles - Molecular assembly - Nitroxide molecules - Nitroxide radicals - Nitroxides - Phase memory - Pulsed electron - Randomly distributed - Spin bath - Spin interaction - Spin-spin - Temperature dependence - Temporal fluctuation - Two-dimensional surface - Unilamellar
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