

The phonon map of molecular qubits

Molecular spin qubits, where quantum information is encoded in electron spin states of paramagnetic systems, offer a unique value proposition for quantum technologies due to their synthetic tunability, scalability, reproducibility and potential for room-temperature operation. While pulsed electron paramagnetic resonance (EPR) is the standard ‘qubit language’ for interrogating molecular qubit coherence – the lifetime of molecular quantum superposition states before they are disrupted by environmental interactions – the microscopic origins of decoherence remains a challenge. A critical bottleneck is spin–phonon coupling (SPC), which drives spin–lattice relaxation (T_1) and limits operation at elevated temperatures.

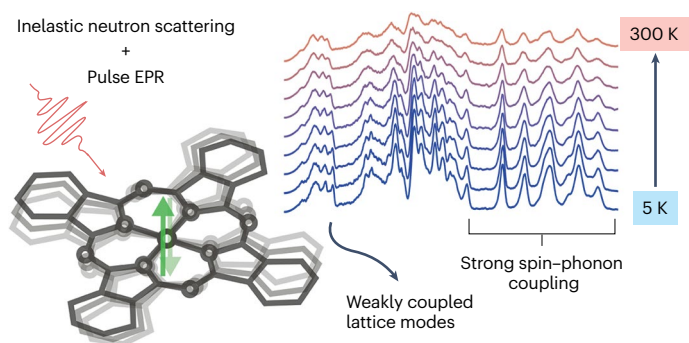
Despite extensive theoretical debate over whether low-energy lattice modes or high-energy optical phonons dominate these relaxation pathways, direct experimental evidence has remained elusive. Writing in the *Journal of the American Chemical Society*, Ryan Hadt and co-workers address this debate by pairing inelastic neutron scattering (INS) with pulsed EPR. This dual-experimental approach provides the first direct experimental quantification of SPC coefficients, offering a rigorous framework to correlate specific vibrational modes with spin relaxation, revealing how precise structural tuning can shield qubits from thermal decoherence.

The researchers used INS to elucidate the phonon spectrum and pulsed EPR spectroscopy to track the temperature-dependent relaxation behaviour, subsequently correlating the two datasets. The chosen qubit systems were the electron spin $S = 1/2$ complexes of CuPc (copper phthalocyanine, D_{4h} symmetry) and saddled CuOEP (copper octaethylporphyrin, adopting a D_{2d} symmetry instead of its standard D_{4h} symmetry), both featuring CuN_4 square planar coordination geometries. From the analysis, the researchers found that in the low-temperature regime below 40 K, relaxation is primarily dominated by low-energy phonons ($\sim 50\text{ cm}^{-1}$), whereas in the high-temperature regime above 40 K, high-energy optical phonons ($\sim 185\text{ cm}^{-1}$) dominate the relaxation process, with coupling strengths nearly three orders of magnitude larger than those in the low-energy and low-temperature regime. The landmark observation is that high-energy phonons couple orders of magnitude more strongly to the spin system than low-energy lattice modes. For example, in CuPc, the high-energy SPC (of $127\text{ }\mu\text{s}^{-1}$) is approximately 1,870 times stronger than the low-energy coupling (at $0.068\text{ }\mu\text{s}^{-1}$). Similarly, in CuOEP, the high-energy coupling ($20\text{ }\mu\text{s}^{-1}$) is roughly 600 times stronger than the low-energy regime ($0.033\text{ }\mu\text{s}^{-1}$). These quantitative results clearly identify high-energy optical phonons as the primary

‘spin coherence limiters’ at elevated temperatures, since stronger spin–phonon coupling accelerates spin relaxation and shortens coherence lifetimes. CuOEP performs better because its distorted D_{2d} geometry, with out-of-plane β -ethyl substitutions, acts as a vibrational damper and stiffens the core environment, raising the energy of relaxation-driving modes and reducing their coupling to the spin centre compared to planar CuPc.

It was previously believed that lower molecular symmetry (such as D_{2d} point groups) causes stronger decoherence because of enhanced vibrational mixing. However, the work by Hadt et al. demonstrates that such a relationship is not always straightforward. By comparing CuPc and CuOEP, the researchers suggest that symmetry breaking can sometimes be beneficial, enabling room-temperature spin coherence in CuOEP. The study also found that intermediate modes between 50 cm^{-1} and 185 cm^{-1} contain phonons that couple only weakly to the spin.

Taken together, this work demonstrates that the combined INS–EPR approach is a highly promising methodology for investigating other molecular qubit systems. Such complementary spectroscopic approaches could provide direct insights into spin–lattice relaxation pathways and enable a deeper microscopic understanding of decoherence mechanisms in next-generation molecular quantum materials.



Kavipriya Thangavel  

National High Magnetic Field Laboratory,
Florida State University, Tallahassee, FL, USA.

✉ e-mail: kthangavel@magnet.fsu.edu

Competing interests

The authors declare no competing interests.

Original article: Lohaus, S. H., Xia, K. T., Cheng, Y. & Hadt, R. G. Experimental quantification of spin–phonon coupling in molecular qubits using inelastic neutron scattering. *J. Am. Chem. Soc.* **148**, 20054–20063 (2026)

Related article: Kang, M. et al. Seeking a quantum advantage with trapped-ion quantum simulations of condensed-phase chemical dynamics. *Nat. Rev. Chem.* **8**, 340–358 (2024)