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Spectroscopy of Nuclear Magnetic Resonance with Single Spin Sensitivity

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Editorial

At the ultimate sensitivity limit of single molecules or single nuclear spins, nuclear magnetic resonance spectroscopy and magnetic resonance imaging necessitate fundamentally novel detection methodologies. One such method is the strong coupling regime, in which interaction between sensor and sample spins dominates all other interactions. This regime allows for the identification of totally unpolarized nuclei, which is previously banned, and goes beyond statistical fluctuations in magnetization. In this paper, we demonstrate robust coupling between an atomic (nitrogen–vacancy) sensor and sample nuclei in order to perform nuclear magnetic resonance on four Si spins. We use the diamond atomic sensor's field gradient in conjunction with compressed sensing to accomplish imaging methods that allow individual nuclei to be located with Angstrom precision. Under ambient conditions, the achieved signal-to-noise ratio enables for single nuclear spin sensitivity in seconds.

Nuclear magnetic resonance (NMR) spectroscopy is a useful method for determining the chemical composition of macromolecules, particularly big proteins. However, the weak contact strength between sample spins and inductive detectors, as well as the low thermal polarisation, limit the sensitivity to large ensembles. The largest magnetic gradient that can be applied further limits the resolution of imaging techniques. Sensor-sample coupling can be improved by miniaturising the detector and bringing it closer to the sample. Magnetic resonance force microscopy and diamond-based magnetometers have recently been used to show NMR on nanoscale ensembles of nuclear spins, enhancing sensitivity by orders of magnitude over the best inductive readout. When noise in the form of magnetic coupling between sample nuclear spins surpasses interaction with the sensor, the sensitivity is traditionally limited to measuring statistical fluctuations in sample magnetization. When coupling between the sensor and the measured nuclei dominates over decoherence (as seen in the strong coupling regime), individual nuclei can be recognised regardless of their polarisation. We achieve such strong coupling by placing a single electronic spin sensor in close proximity to weakly interacting Si nuclei in a silica layer (2 nm).

Additionally, strong coupling allows the atomic sensor's dipolar field to be employed as a field gradient for magnetic resonance imaging, allowing the positions of four single nuclear spins to be observed. The ambient experimental approaches we present represent a significant step toward non-destructive imaging of single biomolecules under physiological settings, as well as detecting nanoscale structure and composition without ensemble averaging. Diamond's nitrogen–vacancy (NV) defect is a unique magnetic sensor. Fluorescence microscopy can probe the NV electronic spin using optically detected magnetic resonance, and dipolar interaction with surrounding spins—either in the diamond or on the surface—allows spin spectroscopy to be performed. Nanoscale magnetic field measurements, bioimaging under ambient settings, and quantum information processing are some of the other uses.

The next section focuses on the possibility of achieving direct flip-flops using unpolarized nuclear spins, that is, regardless of the nuclear spin's initial state. This protocol's sensitivity is critically dependent on the stand-off distance between the NV sensor and target spins, not only because the interaction strength scales inversely with the third power of the separation distance, but also because the signal characteristics change dramatically when the interaction strength between NV and nuclear spins exceeds the coupling between target nuclei. All nuclear spins directly impart phase accumulation on the NV sensor in this strong coupling regime before spin flips between nuclei, which operate to randomise the phase accumulation. When compared to the classical scenario, the result is an improvement in the signal for detecting N nuclei. without the need for sample hyperpolarization The sensitivity we obtain allows us to detect a single unpolarized nuclear spin in less than 10 seconds [1-5].

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