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Microscopic theory of electron spin relaxation in N@C_{60}[1] Z.G. YU, SRI International — Endohedral N@C_{60} exhibits an extremely long electron spin relaxation time and offers a great potential in storing and processing quantum information. Here we present a microscopic theory of electron spin relaxation in N@C_{60}. The theory combines (1) the spin-orbit interaction of N $2p$ electrons, which mixes the ground state ^4S with excited ^2P and ^2D states, and (2) the coupling between the N $2p$ electrons and C_{60} H_g vibrations, which facilitates transitions between ^2P and ^2D states. The spin relaxation occurs via a two-phonon (Raman) process by absorbing a H_g phonon and emitting another at the (approximately) same frequency. The theory consistently explains measured spin relaxation time T_1 and its temperature dependence, and predicts two distinct spin decoherence T_2 constants. In addition, the excellent agreement between theory and experiment suggests a universal importance of the two-phonon Raman process in determining spin relaxation in nanostructures such as quantum dots, where a one-phonon process is ineffectual in flipping electron spins because of the lack of low-energy phonons in nanostructures.

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