$3d$ Electron Quadrupole Moments in Vanadium Oxides

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Vanadium oxides, such as VO_2, V_2O_3, and Magnéli phases $V_nO_{2n\pm1}$, are conventional examples of the Mott transition. The metal-insulator transition in these systems has been recently revisited from a view of orbital physics. We have investigated the obital occupancy in V_2O_3 ($3d^2$), V_5O_9 ($3d^{1.4}$), and V_6O_{13} ($3d^{0.66}$) via the single-crystal ^{51}V NMR measurements. Determination of the hyperfine coupling tensor revealed almost equivalent occupation of a_{1g} and e'_g orbitals, which leads to the vanishing electron quadruple moment, in the paramagnetic metallic state of V_2O_3. We observed the appearence of the quadrupole moment in the low-temperature metallic state under pressure of 2.5 GPa, where the developement of antiferromagnetic correlations was observed in the nuclear spin-lattice and spin-spin relaxation measurements. In contrast, a large quadrupole moment was found in the metallic state of V_5O_9 and V_6O_{13}. We observed the formation of local spin-singlets accompanied by obital order in these two componds. The t_{2g} orbital state changes across the metal-insulator transition, which is manifested in the asymmetric electric field gradient at the ^{51}V nucleus. The significant interplay among the spin, charge, and orbital degrees of freedom in the vanadium oxides is discussed.