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HOMO-1 Contribution in High Harmonic Generation¹ JOSEPH FARRELL, BRIAN MCFARLAND, MARKUS GUEHR, PHILIP BUCKSBAUM, Stanford PULSE Center — High harmonic generation (HHG) proceeds in three steps. First, a part of the electron wave function tunnels out of the valence orbital. Secondly, the liberated electron wave packet accelerates in the laser field and finally coherently recombines with the initially ionized orbital. In molecules the highest occupied molecular orbital (HOMO) is generally thought to be responsible for ionization and recombination. We have obtained experimental evidence that lower lying orbitals are involved in the HHG process of the N_2 molecule. Harmonic signal modulations at different molecular alignment angles display features due to a π_u symmetry and have a higher spectral cut-off than the main HHG spectrum from the σ_g – symmetric HOMO. This suggests HHG from the HOMO-1 orbital.

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