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In this work, we take into account our recent studies on C_3 where we used perturbation-facilitated two-color resonant four-wave mixing spectroscopy to access the (dark) triplet manifold from the singlet ground state *via* “gate-way” levels (i.e. singlet-triplet mixed levels) [1] and the deperturbation investigation of the $d^3\Pi_g$, $v=6$ level of the Swan band that unveiled the presence of the lowest quintet state ($^5\Pi_g$) of C_2 [2].

We report on perturbation-facilitated optical-optical double-resonance experiments to access the first excited quintet state of C_2 *via* “gate-way states” in the perturbed $d^3\Pi_g$, $v=6$. The newly found $^5\Pi_u$ state is characterized at rotational resolution by performing a least-squares fit of the observed transitions to a $^5\Pi_u$ - $^5\Pi_g$ Hamiltonian. The work represents a rare case of a successful analysis of a quintet manifold of a molecule exhibiting a singlet ground state ($^1\Sigma_g^+$).

[1] Y. Sych, P. Bornhauser, G. Knopp, Y. Liu, T. Gerber, R. Marquardt, and P.P. Radi, *J. Chem. Phys.* 139, 154203 (2013)

[2] P. Bornhauser, Y. Sych, G. Knopp, T. Gerber, and P.P. Radi, *J. Chem. Phys.* 134, 044302 (2011)

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