# Femtosecond dynamics of correlated many-body states in $C_{60}$ fullerenes

**Sergey Usenko[1], Michael Schüler[2], Armin Azima[3,4,5], Markus Jakob[1,5], Leslie L. Lazzarino[3], Yaroslav Pavlyukh[2,6], Andreas Przystawik[1], Markus Drescher[3,4,5], Tim Laarmann[1,5], Jamal Berakdar[2]**

[1] Deutsches Elektronen-Synchrotron DESY, D-22607 Hamburg, Germany
[2] Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, D-06099 Halle, Germany
[3] Department of Physics, University of Hamburg, D-22761 Hamburg, Germany
[4] Center for Free-Electron Laser Science CFEL, DESY, D-22607 Hamburg, Germany
[5] The Hamburg Centre for Ultrafast Imaging CUI, D-22761 Hamburg, Germany
[6] Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, D-67653 Kaiserslautern, Germany

**Synopsis** In this joint theoretical and experimental work we investigate the population and decay dynamics of excited states of the $C_{60}$ molecule by time-resolved two-photon photoemission. We map out how the thermally excited vibrational degrees of freedom lead to a transient redistribution of the photo-excited states. This includes the super-atom molecular orbitals (SAMOs), which are of great interest currently. The measured lifetimes are in line with full-fledged first-principle calculations.

Fullerene complexes hold great promises for molecular junctions or organic solar cells. In particular, extended, nearly atom-like orbitals, the super-atom molecular orbitals (SAMOs) represent – in view of their long lifetimes – potentially very robust transport channels for nano-sized devices. Insights in the control and dynamics of charge currents and their dissipation can be gained on the femtosecond time scale. To this end, ultrafast optical spectroscopy has become a major asset.

In this work [1], we investigate the excitation and dissipation dynamics of correlated states of the $C_{60}$ molecule in the gas phase. Based on first-principle time-dependent density-functional theory (TDDFT) for the excited states of the molecule, a time-resolved two-photon photoemission (2PPE) setup is employed to map out the dynamics of the excited states. The experiments are further supported by time-dependent simulations of the laser-induced dynamics based on *ab initio* calculations of the coupling of the electronic degrees of freedom to the thermally excited vibrations.

Fig. 1(a) shows the simulated dynamics induced by the pump pulse as in the experiment. After initially excited an optically active bright state (BS), the vibrational relaxation and thermalization leads to a transient population of several dark states (DSs), including the SAMOs. The theoretical calculations further allow for inferring the lifetime of the laser-excited states and their manifestation on the pump-probe experiment. The result is compared to the experiment in Fig. 1(b). The decay behavior of the transient 2PPE signal matches the theory very well and thus identifies the vibrational degrees of freedom as major source of the dissipation.

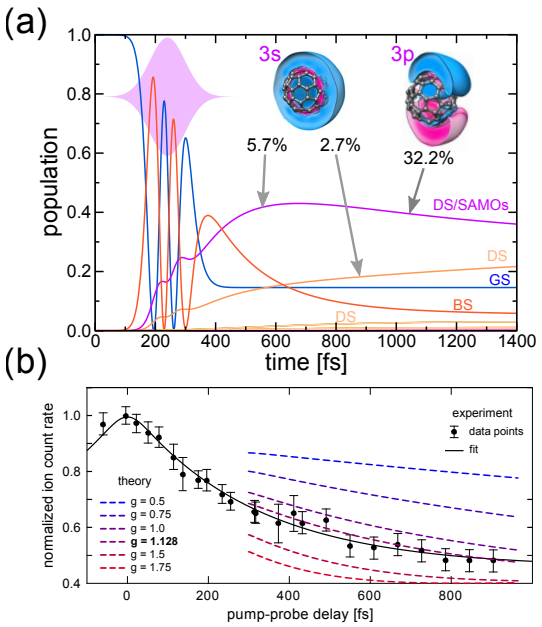


**Figure 1**. (a) Population dynamics induced by the pump pulse (sketched in the background). The insets show the weight of the SAMOs in the dominantly populated states. (b) Normalized $C_{60}^{+}$ counts as a function of pump-probe delay. The signal derived from theory are compared to the experimental results for different scaling factors $g$ of the electron-vibron interaction strength in the limit of long pump-probe delays. The bold value indicates the best agreement between theory and experiment.

## References

[1] S. Usenko, M. Schüler *et al.* 2015 *New. J. Phys.* **18** 113055

[1]E-mail: michael.schueler@physik.uni-halle.de