Nonlinear Optics and Quantum Tunneling in Plasmonic Nanocavities

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Plasmonic molecular nanojunctions exhibit opto-mechanical coupling at the nanoscale, enabling intertwined optical, vibrational and electronic phenomena. In this work, we demonstrate plasmon-mediated phonon pumping, driven by inelastic electron hopping in conductive molecules, which results in strong Raman nonlinearity at the light intensities almost three orders of magnitude lower than in the conventional opto-mechanical systems and up to four-fold enhancement of the effective Raman polarizability due to vibrational electron-phonon coupling. We also developed a microscopic framework of opto-mechanical electron-phonon coupling in molecular nanojunctions based on the Marcus electron hopping. Systematically varying electrical conductance of the molecules in the junction and laser intensity, we observed the transition between a photo-assisted tunneling regime and an electron hopping process. Our findings provide a microscopic description for vibrational, optical, and electronic phenomena in plasmonic nanocavities important for efficient phonon lasing, representing the first attempt to exploit conductive molecules as quantum-mechanical oscillators.

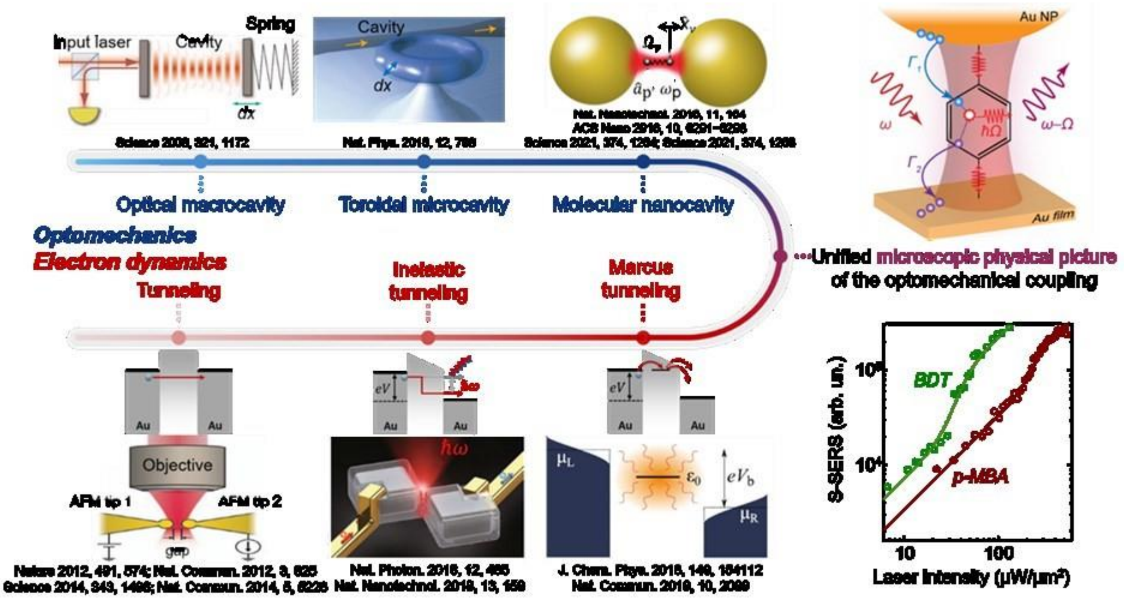

Fig. 1 Schematic overview of the research background and our new concept. The blue axis highlights key milestones in the development of optomechanics, with references to representative works. The red axis outlines significant advancements in the study of electron tunneling in nanoscale gaps, accompanied by notable references. On the right, we present the mechanistic illustration underlying this work, along with our main experimental data and corresponding theoretical fitting.

References

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