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Terahertz Photonics of Quasi-One-Dimensional Carbon Nanostructures

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The technology for synthesizing carbynes, also known as linear acetylenic or polyynic carbons, has experienced significant advancements over the past few years [1]. Stable long aligned chains of carbynes are now being successfully deposited on substrates, and we have recently shown [2] that highly aligned arrays of carbynes terminated by gold clusters produce luminescence highly sensitive to the polarization of the exciting light. This paves the way for a new class of ultimately thin polarization-sensitive emitters to be used in future integrated quantum photonics devices. Another important characteristic of long polyyne chains, which feature two alternating non-equal bonds, is the presence of mid-gap edge states that are topologically protected. In a finite-length chain, these two edge states form an even and odd combination, with the energy gap proportional to the overlap of the edge states due to tunneling. These split states of different parity support strong dipole transitions. Our research [3] has demonstrated that, for carbyne chains of a certain length the energy separation between the HOMO and LUMO molecular orbitals formed by the edge states corresponds to the THz frequency range. This HOMO-LUMO energy gap can be tuned by external electric field. Additionally, there are several other allowed optical transitions in this system that can be used to maintain the inversion of population required for THz lasing.

Another recent achievement in nanocarbon technology is a demonstration of controlled synthesis of cyclocarbons, in particular cyclo[18]carbon allotrope [4]. The properties of cyclocarbons in an external electric field differ drastically depending on the parity of the number of dimers in a polyyne ring. This is a direct consequence of breaking the inversion symmetry in a ring consisting of an odd number of dimers, including the famous C_{18}. Our estimates [5] show that adding just one extra carbon dimer to C_{16} is equivalent to placing this molecule in an external magnetic field of 104 T. For an odd-dimer cyclocarbon, as a result of the inversion symmetry absence, an experimentally attainable electric field should open a tunable gap between otherwise degenerate states leading to two states with allowed dipole transitions between them in the THz range. A population inversion can be achieved again using optical pumping.

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1. S. Kutrovskaya, A. Osipov, S. Baryshev, A. Zasedatelev, V. Samyshkin, S. Demirchyan, O. Pulci, D. Grassano, L. Gontrani, R. R. Hartmann, M. E. Portnoi, A. Kucherik, P. G. Lagoudakis, and A. V. Kavokin, "Excitonic fine structure in emission of linear carbon chains," *Nano Lett.* **20**, 9, July 2020, pp. 6502-6509, doi: 10.1021/acs.nanolett.0c02244.

2. A. Kucherik, A. Osipov, V. Samyshkin, R. R. Hartmann, A. V. Povolotskiy, M. E. Portnoi, "Polarization-Sensitive Photoluminescence from Aligned Carbon Chains Terminated by Gold Clusters," *Phys. Rev. Lett.,* **132**, 5, February 2024, Art. No 056902 , doi: 10.1103/PhysRevLett.132.056902.

3. R. R. Hartmann, S. Kutrovskaya, A. Kucherik, A. V. Kavokin, and M. E. Portnoi, "Terahertz transitions in finite carbon chains," *Physical Review Research* **3**, 3, September 2021, Art. No 033202, doi: 10.1103/PhysRevResearch.3.033202.

4. K. Kaiser, L. M. Scriven, F. Schulz, P. Gawel, L. Gross, H. L. Anderson, "An sp-hybridized molecular carbon allotrope, cyclo[18]carbon. *Science* **365**, 6459, September 2019, pp. 1299–1301, doi: 10.1126/science.aay1914.

5. R. A. Ng, M. E. Portnoi, and R. R. Hartmann, "Tuning terahertz transitions in cyclo[n]carbon rings," *Phys. Rev. B*, **106**, 4, July 2022, Art. No L041403, doi: 10.1103/PhysRevB.106.L041403.