

# Study finds the origin of 'odd-even' effects

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(Phys.org)—The most intriguing property of nano-scale organic devices is their tunability. Their properties can be tweaked by changing the organic structure. There have been several studies exploring the organic structure of alkythiolate self-assembled monolayers (SAMs) anchored to a gold or silver electrode and contacted with a top-electrode, creating a metal—SAM—metal junction. Studies have shown that an odd number of carbons demonstrate a different charge transfer compared to an even number of carbons, a property known as the "odd-even" effect. While this effect has something to do with the junction point near the top electrode, the reason this occurs is still unclear because it is difficult to separate molecular effects from interface effects.

In a paper recently published in the *Journal of the American Chemical Society*, Li Jiang, C.S. Suchand Sangeeth, and Christian A. Nijhuis of the National University of Singapore investigated the cause of the odd-even effect in n-alkylthiolates of varying lengths on a silver electrode and contacted by a GaOx/EGaIn top-electrode using impedance spectroscopy and statistical analysis. This technique allowed them to identify the molecular and interface contributions to the "odd-even" effect.

In the silver n-alkylthiolate system, an odd number of carbons in the alkyl chain impedes the charge more than an even number of carbons. This likely has to do with the alkyl chain conformation, or, more specifically, how it is twisted. However, the typical way to measure current measures total current across the junction and does not parse out how molecular effects and various interface effects contribute to the odd-even effect. Jiang, et al. report the use of both AC and DC methods,

including the use of impedance spectroscopy as they have used in previous studies, to determine the molecule-electrode contact resistance ( $R_c$ ) and the resistance ( $R_{SAM}$ ) and capacitance ( $C_{SAM}$ ) of the SAMs.

Jiang, et al. obtained a large amount of data, approximately 300 current measurements for each type of junction spanning a potential of  $\pm 0.5V$ . This amounted to a total of 5,385 traces obtained from 342 junctions. This amount of data allowed them to conduct a robust statistical study. They related current across the junction to the number of carbon atoms. Then they used impedance spectroscopy on those junctions made from  $n = 6$  to 18 carbons. Odd SAMs impede charge transport more than even SAMs, and furthermore, this contribution is predominantly due to the SAM's resistance and not from SAM-electrode contact resistance.

SAM resistance is dependent upon the SAM-electrode interaction and SAM packing. SAM packing determines the tunneling barrier height. Tunneling barrier height is due to specific molecular and interface effects.

The odd-even effect was most apparent in SAMs with  $n = 2$  to 18 carbons and the magnitude of the odd-even effect increases with increasing  $n$ . Furthermore, the conformation of the alkyl chain, i.e., the twist angle of the SAM, likely contributes to the tunnel barrier height. Overall, it is intrinsic properties of the SAMs that produce the odd-even effect, and therefore, these SAMs can be tweaked to tailor the properties of a molecular device.

**More information:** "The origin of the odd-even effect in the tunneling rates across EGaIn Junctions with SAMs of n-alkanethiols", Li Jiang, C.S. Suchand Sangeeth, and Christian A. Nijhuis, *J. Am. Chem. Soc.* DOI: [10.1021/jacs.5b0576](https://doi.org/10.1021/jacs.5b0576)

## Abstract

Odd-even effects in molecular junctions with self-assembled monolayers (SAMs) of n-alkanethiolates have been rarely observed. It is challenging to pinpoint the origin of odd-even effects and address the following question: are the odd-even effects an interface effect, caused by the intrinsic properties of the SAMs, or a combination of both? This paper describes the odd-even effects in SAM-based tunnel junctions of the form AgA-TS-SCn//GaOx/EGaIn junctions with a large range of molecular lengths ( $n = 2$  to 18) that are characterized both by AC and DC methods along with a detailed statistical analysis of the data. This combination of techniques allowed us to separate interface effects from the contributions of the SAMs and to show that the odd-even effect observed in the value of  $J$  obtained by DC-methods are caused by the intrinsic properties of the SAMs. Impedance spectroscopy (an AC technique) allowed us to analyze the SAM resistance ( $RSAM$ ), SAM capacitance ( $CSAM$ ), and contact resistance, within the junctions separately. We found clear odd-even effects in the values of both  $RSAM$  and  $CSAM$ , but the odd-even effect in contact resistance is very weak (and thus is thus not responsible for the observed odd-even effect in the current densities obtained by  $J(V)$  measurements). Therefore, the odd-even effects in AgA-TS-SCn//GaOx/EGaIn junctions are contributed by the properties of the SAMs and SAM-electrode interactions which both determine the shape of the tunneling barrier.

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