Supplementary Information for:

Reassessing destructive quantum interference in azulene-based devices

Supplementary Table S1: Optimized bond lengths of each system (Å), molecular length (Å) and the zero-bias conductance (μS) for the two-probe systems.

Junction	C≡C	C—C	S—S	*G* (μS)
2,6Az-I			8.80	15.46
2,6Az-II	1.24	1.41	13.94	9.79
2,6Az-III		1.47	17.48	0.75
2,6Az-IV	1.23	1.38	22.36	5.42
4,7Az-I			6.74	13.17
4,7Az-II	1.24	1.42	11.75	10.73
4,7Az-III		1.47	15.06	0.12
4,7Az-IV	1.21	1.42	20.03	0.50
1,3Az-I			5.65	10.85
1,3Az-II	1.24	1.39	11.75	10.81
1,3Az-III		1.47	18.56	0.07
1,3Az-IV	1.22	1.42	18.56	0.07
5,7Az-I			5.40	3.48
5,7Az-II	1.23		8.37	2.27
5,7Az-III		1.40	12.10	0.01
5,7Az-IV	1.21	1.42	16.03	0.01

Supplementary Note 1 | Sample Input Files for Calculations.

1) Siesta

Version: 3.2-pl-5

M1 SCF input file:

```
# ------------------------------------------------------------------------
# Name and Label
# ------------------------------------------------------------------------
SystemName          2,6Az-I
SystemLabel         2,6Az-I.file
#------------------------------------------------------------------------
```

```
# Lattice
#-------------------------------------------------------------------------
LatticeConstant            1.00 Ang
%block LatticeVectors
4.44417          7.69752          0.000000
8.88833          0.000000         0.000000
 0.000000        0.000000        33.9459508971
%endblock LatticeVectors
# -------------------------------------------------------------------------
# Species and Atoms
# -------------------------------------------------------------------------
NumberOfSpecies       4
NumberOfAtoms         54
%block ChemicalSpeciesLabel
  1   pseudos/H-gga
  6   pseudos/C-gga
  16  pseudos/S-gga
  79  pseudos/Au-gga
%endblock ChemicalSpeciesLabel
# -------------------------------------------------------------------------
# Atomic Coordinates
# -------------------------------------------------------------------------
AtomicCoordinatesFormat Ang
%block AtomicCoordinatesAndAtomicSpecies
   2.962777000004       5.131681000003       8.466837000005       4
   1.481389             2.565840999998       8.466837000005       4
   0.                   0.                   8.466837000005       4
   5.925554999997       5.131681000003       8.466837000005       4
   4.444166000003       2.565840999998       8.466837000005       4
   2.962777000003       0.                   8.466837000005       4
   8.888332             5.131681000003       8.466837000005       4
```

7.406943999996	2.565840999998	8.466837000005	4
5.925554999996	0.	8.466837000005	4
4.444166000005	5.986961000002	10.885935000022	4
2.962777000001	3.421120999997	10.885935000022	4
1.481389	0.855279999999	10.885935000022	4
7.406943999998	5.986961000002	10.885935000022	4
5.925554999994	3.421120999997	10.885935000022	4
4.444166000003	0.855279999999	10.885935000022	4
10.369721	5.986961000002	10.885935000022	4
8.888331999997	3.421120999997	10.885935000022	4
7.406943999996	0.855279999999	10.885935000022	4
7.406943666669	4.276401000004	12.573935000014	3
5.745433813692	3.112999157519	14.300929231871	1
9.221429628163	5.546919098533	14.319152453981	1
7.481638860727	4.328703143283	14.363250359839	2
6.443210212706	3.60158750191	14.977142965564	2
8.515815741791	5.052841665079	14.981595535873	2
6.178258052876	3.416065983416	16.332047042944	2
8.771023835745	5.231540314498	16.342309363745	2
5.304136504014	2.803999423185	16.549732186211	1
9.644206989952	5.84294980365	16.566561748227	1
6.862866117564	3.895433759905	17.452409562586	2
8.081771455128	4.748920552653	17.455597511565	2
6.524490386756	3.658500498288	18.793193162151	2
8.414660496399	4.982011992503	18.800987161222	2
5.683899378967	3.069912278071	19.138808802918	1
9.25442510153	5.570021559291	19.150385135747	1
7.468454678749	4.319471478735	19.609132626638	2
7.406943666669	4.276401000001	21.37201589707	3
4.444166000005	5.986960999999	23.060015897062	4
2.962777000001	3.421120999994	23.060015897062	4

```
 1.481389              0.855279999997    23.060015897062    4
 7.406943999998        5.986960999999    23.060015897062    4
 5.925554999994        3.421120999994    23.060015897062    4
 4.444166000003        0.855279999997    23.060015897062    4
10.369721              5.986960999999    23.060015897062    4
 8.888331999997        3.421120999994    23.060015897062    4
 7.406943999996        0.855279999997    23.060015897062    4
 2.962777000003        6.842240999999    25.479113897079    4
 1.481389000003        4.276401000001    25.479113897079    4
-0.000000000001        1.710559999996    25.479113897079    4
 5.925555000005        6.842240999999    25.479113897079    4
 4.444166000006        4.276401000001    25.479113897079    4
 2.962777000001        1.710559999996    25.479113897079    4
 8.888331999999        6.842240999999    25.479113897079    4
 7.406943999999        4.276401000001    25.479113897079    4
 5.925554999995        1.710559999996    25.479113897079    4
%endblock AtomicCoordinatesAndAtomicSpecies
#----------------------------------------
# Calculator
# ----------------------------------------
#----------------------------------------
# Exchange-Correlation
#----------------------------------------
SolutionMethod          diagon
XC.functional           GGA
XC.authors              PBE
PAO.BasisSize           DZP
MeshCutoff              300.0 Ry
DM.Tolerance            1.d-5
%block kgrid_Monkhorst_Pack
 6   0   0      0.0
```

```
0   6   0      0.0

0   0   7      0.0

%endblock kgrid_Monkhorst_Pack
```

2) Nanodcal

Electrode SCF input file:

```
system.name = ' 2,6Az-I '

system.spinType = ' NoSpin '

system.orbitalType = ' DZP '

system.atomCoordinateFormat = ' cartesian '

system.atomFile = ' ./2,6Az-I-elec.xyz '

system.centralCellVectors =' [[4.44417  7.69752 0.0]',[8.88833  0.0  0.0]',[0.0 0.0 7.25728999999]']  '

calculation.name = ' scf '

calculation.SCF.startingMode = ' H '

calculation.SCF.mixingMode = ' H '

calculation.SCF.donatorObject = ' NanodcalObject.mat '

calculation.control.precision = 'high'

calculation.k_spacegrids.number = [6,6,100]

calculation.xcFunctional.Type = '  GGA_PBE96 '
```

Central region SCF input file:

```
system.name = ' 2,6Az-I '

system.spinType = ' NoSpin '

system.orbitalType = ' DZP '

system.centralCellVectors =  '[[8.888330 0.0 0.0]',[4.444170  7.697520 0.0]',[0.0 0.0 45.102908]'] '

system.atomFile = ' 2,6Az-I.xyz '

calculation.name = ' scf '

calculation.SCF.donatorObject = ' NanodcalObject.mat '

system.numberOfLeads = 2

system.typeOfLead1 = left

system.objectOfLead1 = ../2,6Az-I_elec/NanodcalObject.mat

system.typeOfLead2 = right

system.objectOfLead2 = ../2,6Az-I_elec/NanodcalObject.mat

calculation.control.precision = ' high '

calculation.k_spacegrids.number = [6,6,1]

calculation.xcFunctional.Type = GGA_PBE96
```