Soft Semiconductor Devices

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Our group works with soft semiconductors. These are materials comprised of molecules held together by weak *van der Waals* bonds. In comparison, the atoms in conventional semiconductors are held together by strong covalent bonds. Weak intermolecular bonds offer a tradeoff to engineers. Their disadvantage is an enhancement of disorder and charge localization, yielding relatively poor charge transport properties.

But the advantage of soft semiconductors is that they are uniquely suited for large area electronics such as video displays, and solar cells. In contrast with the painstaking growth requirements of conventional semiconductors, films of soft semiconductors are readily deposited on a variety of materials at room temperature. Most importantly, the optical properties of the molecules within a soft semiconductor are relatively immune to structural defects and disorder in the bulk. Thus, soft semiconductors are tolerant of the defects that inevitably occur in the fabrication of large area applications.
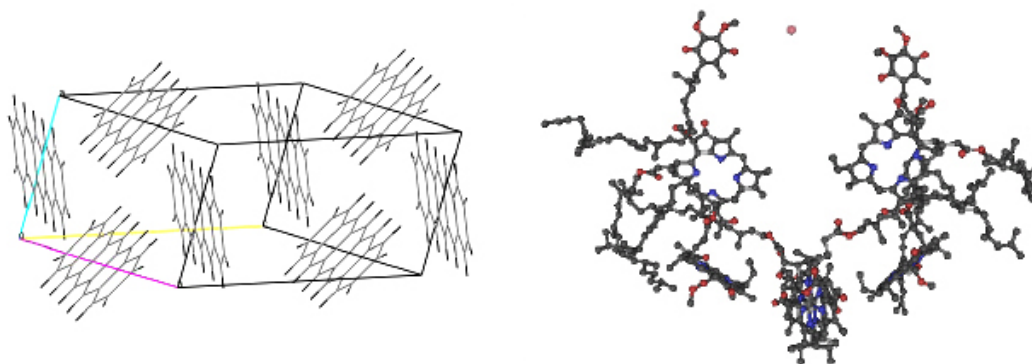

Figure 1: Two examples of van der Waals bonded semiconductors. **(Left)** The structure of a molecular crystal of pentacene. Pentacene has a hole mobility of approximately 2 cm2/Vs and is widely used in organic electronics. Data is from Mattheus, et al. Acta Crystallographica C. 57, 939 (2001). **(Right)** The structure of the photosynthetic reaction center of Rhodobacter sphaeroides. Evolved over two billion years, this molecular circuit is the backbone of photosynthesis. The protein scaffold has been removed for clarity. Data is from Ermler, et al. Structure 2, 925 (1994).

This is a young field, with much promise and significant challenges. In our work, we have successfully integrated complex structures from photosynthesis with solid-state electronics. We also work in two of the major controversies in thin film soft semiconductors: the fundamental efficiency limit of organic light emitting devices, where we have questioned accepted models of excited-state formation; and charge injection, where we have developed a theory centered on structural and energetic disorder in these soft materials.

1.   Vertical Dye Alignment in Luminescent Solar Concentrators for Improved Trapping Efficiencies

Sponsor
Center for Excitonics, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award Number DE-SC0001088

Project Staff
C. L. Mulder, P. D. Reusswig, A. M. Velázquez , H. Kim, C. Rotschild, M. A. Baldo

Luminescent Solar Concentrators (LSCs) aim to reduce the cost of solar electricity by using an inexpensive collector to concentrate solar radiation without mechanical tracking (Fig. 1a) [1]. Ideally, the dyes re-emit the absorbed light into waveguide modes that are coupled to solar cells (black arrows). But some photons are always lost, re-emitted through the face of the LSC and coupled out of the waveguide (grey arrows). In this work, we improve the fundamental efficiency limit of an LSC by controlling the orientation of dye molecules using a liquid crystalline host. Conventional LSCs employ randomly aligned dyes that collectively emit photons isotropically; see Fig. 1a. This yields a trapping efficiency of $\eta_{trap} \sim 0.75$ for a waveguide refractive index of $n_S = 1.5$. Instead, it is possible to align the dyes vertically so their transition dipoles couple more strongly into the waveguide, as illustrated in Fig. 1b. We employ a homeotropic liquid crystal matrix to align rod-shaped dye molecules perpendicular to the waveguide, as illustrated in Fig. 1c. Orienting the molecules at a right angle to the surface reduces the absorption of perpendicular incident radiation. Consequently, we employ an optical diffuser above the LSC to scatter incident light.

We have enhanced the overall performance of LSCs by 16% by aligning the dipole moment of dye molecules perpendicularly to the waveguide. The improvement is due to an increase in trapping efficiency. We measure η_{trap} = 81% for an LSC employing vertically aligned dye molecules compared to η_{trap} = 66% for an LSC based on randomly oriented dye molecules (see Fig. 2). The increase is consistent with theory that models the system using Hertzian dipoles embedded in a waveguide. This theory also predicts small but significant changes to the commonly-cited trapping efficiency of LSCs employing isotropic dipoles [2].
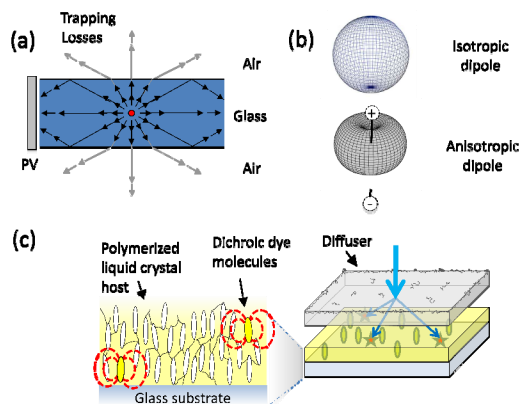

Figure 1: (a) A schematic representation of a luminescent solar concentrator. The trapping efficiency is defined as the fraction of photons emitted from the edge versus photons emitted from the face and edge combined.   (b) The emission profile of isotropic dipoles and a linearly aligned, anisotropic dipole. The absorption and emission profile of isotropic dipoles is uniform, while the anisotropic dipole emission is characterized by a $\sin^2\theta$ profile. (c) In order to improve the performance of LSCs, we align rod-shaped dichroic dye molecules perpendicular to the waveguide. An external diffuser is used to correct for the reduced ability to absorb light incident perpendicular to the waveguide.
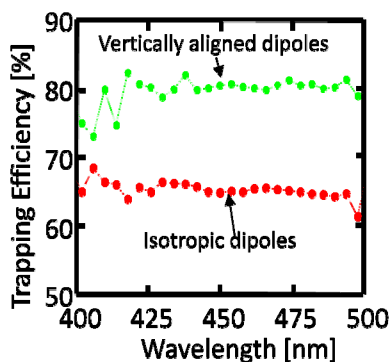
Figure 2: The measured trapping efficiency, η_{trap} of the isotropically aligned LSCs (red dots) and the vertically aligned, homeotropic, LSCs (green dots). This measured η_{trap} is the ratio between the measured number of photons emitted from the edge and the total number of emitted photons from face and edges.

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[1]   J.S. Batchelder, A.H. Zewail, and T. Cole , "Luminescent solar concentrators. 2: Experimental and theoretical analysis of their possible efficiencies", *Applied Optics*, vol. 20, pp. 3733-3754, Nov. 1981**.**

[2]   C. L. Mulder, P. D. Reusswig, A. M. Velázquez, H. Kim, C. Rotschild, and M. A. Baldo, "Dye alignment in luminescent solar concentrators: I. Vertical alignment for improved waveguide coupling", *Optics Express*, accepted for publication**.**

2. Horizontal Dye Alignment in Luminescent Solar Concentrators for Energy Harvesting in Displays

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Most modern displays enhance their contrast ratio using combinations of linear polarizers to absorb and dissipate incident light. The polarizers also absorb more than 50% of the light emitted from the backlight of the display, making them an extremely inefficient component of the modern displays. Thanks to their inherently absorptive surfaces, and their ever-increasing sizes in televisions and mobile electronics, displays are interesting candidates for energy harvesting. In this project, we test a new energy-harvesting strategy in which the purely absorptive polarizers in conventional displays are replaced with two linearly polarized luminescent concentrators (LP-LSCs) that channel the energy of absorbed photons to photovoltaics at the edge of the display.

Conventional polarizers are based on aligned molecules that are highly dichroic in their optical properties, i.e. they absorb light strongly when the E-field is oriented along their long axis, while being relatively transparent to polarizations perpendicular to its natural dipole moment. Our idea is to introduce light-emitting molecules into the matrix of the polarizer so that absorbed photons are not merely dissipated as heat, but are re-emitted within a part of the polarizer that acts as a waveguide (see Fig 1a), just like in a conventional Luminescent Solar Concentrator [1]. These photons travel to the edges of the waveguide by total internal reflection, where they are collected by solar cells. This concept allows the photovoltaics to be located in the frame of the display, which minimizes their area, while leaving the entire front surface available for the display. We create such a linearly polarized LSC (LP-LSC) by using a polymerizable liquid crystal matrix as a host so as to linearly align a rod shaped dye molecule, such as Coumarin 6, in the plane of the substrate (Fig. 1b). Proof of concept experiments show that up to 38% of the photons polarized on the long axis of the Coumarin 6 dye molecules can be coupled to the edge of the device for an LP-LSC with an order parameter of 0.52 (Fig. 2) [2].
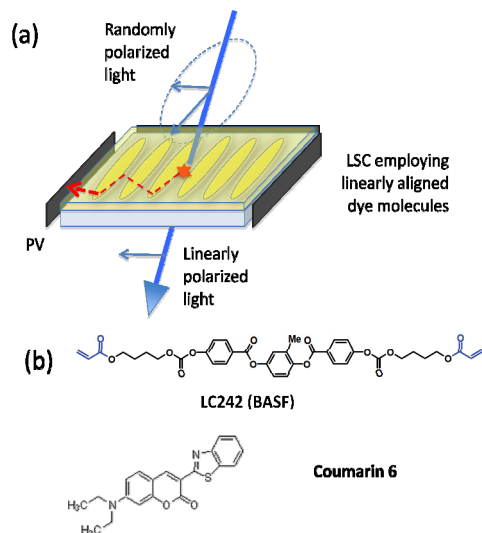
Figure 1: (a) A schematic representation of linearly polarized luminescent solar concentrators (LSC) for energy-harvesting in displays. (b) Chemical structures of the materials used for the linearly polarized LSC studies. The liquid crystal host material used is Paliocolor LC242. The dichroic dye molecules is 3-(2-Benzothiazolyl)-N,N-diethylumbelliferylamine (Coumarin 6).
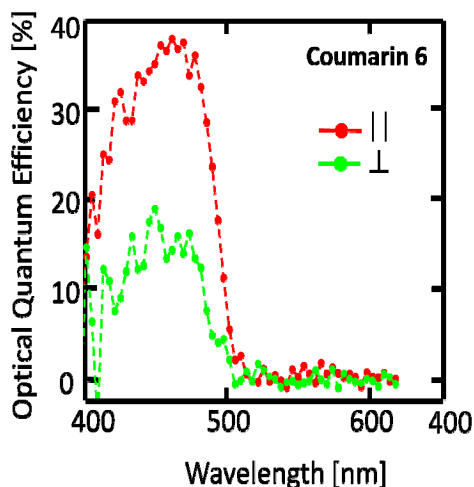
Figure 2: The optical quantum efficiency, defined as the fraction of the photons incident on the face of the luminescent concentrator that is emitted from the edge for a linearly aligned system. Only 70% of the incident photons were absorbed for this device.

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3. Organic Excitonic Transistors

Sponsors

Project Staff

J. Lee, C. L. Mulder, H. Mendoza, K. Milaninia, B. Cord, M.A. Baldo

Excitons, bound pairs of electrons and holes, mediate the interconversion of charges and photons and thus can be used for an efficient interconnect between electronic circuits and optical communication. The ability to guide excitons in space can lead to an excitonic switch that directly routes an optical signal. Recently, High et al. demonstrated an excitonic transistor using indirect excitons formed in AlGaAs/GaAs coupled quantum wells at a temperature of 1.4K [1]. The much larger binding energy of excitons in organic semiconductors could enable excitonic transistors at room temperature. Furthermore, by exploiting spin-disallowed transitions in organic materials, room temperature excitons can last up to milliseconds, more than sufficient to enable exciton propagation over large distances and the operation of sample circuits.

In this work we aim to demonstrate an exciton transistor based on organic semiconductors that can operate at room temperature. Excitons have a dipole moment with a defined electron-hole spacing. The

exciton energy can be controlled by coupling external electric fields with the exciton dipole moment. We propose to guide excitons using the energy gradient determined by external electric fields (See Figure 1). In order to operate this optical switch, excitons are required to diffuse at least roughly over the wavelength of light to overcome the diffraction limit. A triplet exciton, an excited state with a total spin of 1, has a long diffusion length since its recombination is disallowed by spin. Indeed, anthracene crystals show a triplet diffusion length of ~20μm at room temperature. We grow anthracene crystals inside a cavity by melting process, expecting to achieve a uniform and defect-free crystal for efficient exciton diffusion. We expect that this work will open a promising route toward the spatial manipulation of excitons in organic semiconductors.
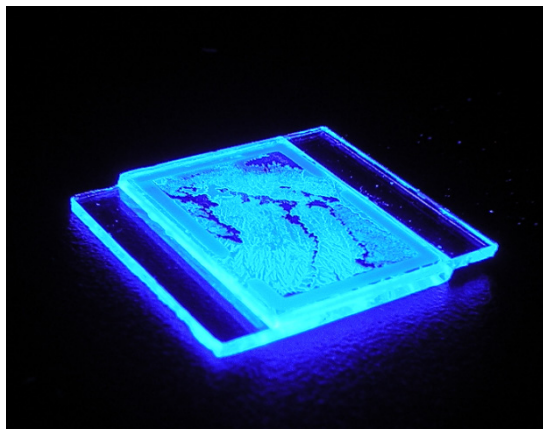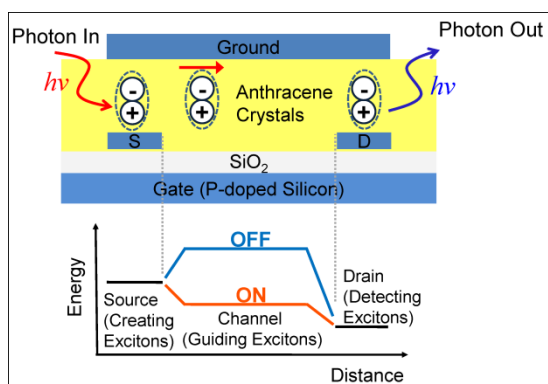

Figure 1: The device structure of the organic excitonic transistor and its energy profile under the operating condition. We control excitonic fluxes by modulating the gate bias and, thus, the potential barrier. The top electrode is transparent, enabling the exciton creation and detection by incoming and outgoing photons.

Figure 2: An anthracene crystal grown inside a cavity sandwiched between two glass substrates. The sample is luminescing blue light under UV excitation.

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4. Singlet Exciton Fission in Organic Solar Cells

Sponsor

Project Staff
Priya Jadhav, Jason Sussman and Marc Baldo

The efficiency of solar cells is controlled by the energy gap of the constituent materials. The 'single junction limit' is imposed because photons with energy higher than the gap lose energy in thermalization and those lower than the gap cannot be absorbed.[ref Schockley Quiesser] Tandem cells work around this by having more than one photovoltaic structure in parallel absorbing different parts of the solar spectrum. But tandem cells are not economical in every material system. So in this project we use singlet exciton fission to potentially surpass the single junction efficiency limit.

Some organic materials, like the acenes: anthracene, tetracene and pentacene, exhibit singlet fission[1], in which a high energy singlet exciton spontaneously decomposes into two low energy triplets. We are investigating the novel device structure shown in Fig. 1. It uses tetracene as a donor to generate high energy singlets in the l = 450-550nm region, which then split into two lower energy triplets, potentially doubling the photocurrent in this part of the spectrum. The presence of singlet fission in tetracene is confirmed by the magnetic field dependence of the photocurrent (Fig. 2). Harvesting of solar energy at longer wavelengths is performed by CuPC (copper phthalocyanine). Thus, this structure is capable of higher efficiencies.
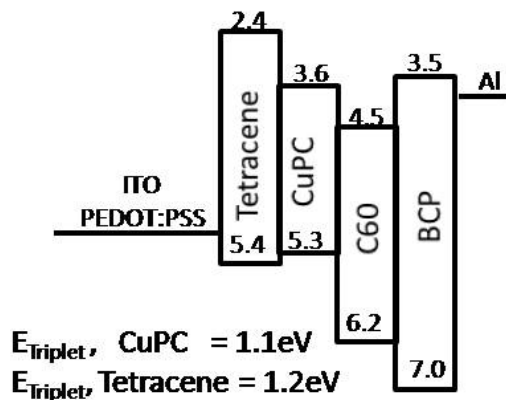

Figure. 1: Device structure and energy levels. The CuPC triplet is slightly lower in energy than the tetracene triplet, allowing the triplets formed in tetracene to reach the interface. BCP prevents exciton quenching at the cathode.
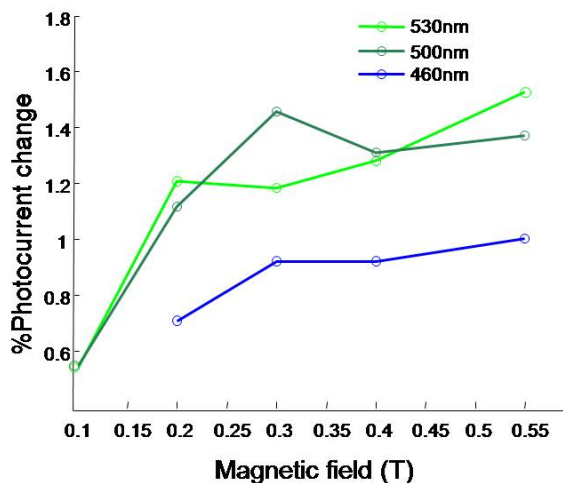

Figure. 2: Change in photocurrent with magnetic field as a function of excitation wavelength. This would be more compelling no change was seen at red wavelengths.

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5. Glass Based Luminescent Solar Concentrator

Sponsor
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Project Staff
P.D. Reusswig, N. Thompson, H. Tuller, and M.A. Baldo

Photovoltaic solar concentrators aim to increase the electrical power obtained from solar cells. Conventional solar concentrators track the sun to generate high optical intensities, often by using large mobile mirrors that are expensive to deploy and maintain. Solar cells at the focal point of the mirrors must be cooled and the entire assembly wastes space around the perimeter to avoid shadowing neighboring concentrators. High optical concentrations without excess heating in a stationary system can be achieved with a luminescent solar concentrator (LSC) [1]. The LSC consist of a dye dispersed in a transparent waveguide. Incident light is absorbed by the dye and then reemitted into a waveguide mode. The energy difference between absorption and emission prevents reabsorption of light by the dye, isolating the concentrated photon population in the waveguide. Unfortunately, the performance of LSCs has been limited by two factors: self-absorption losses and a scarcity of dyes that absorb and emit efficiently in the infrared region to be coupled to silicon solar cells. We have previously made significant progress on the problem of self-absorption losses [2]. Now we address operation in the infrared region. Neodymium (Nd^{3+}) and Ytterbium (Yb^{3+}) are nearly the optimal infrared LSC material: inexpensive, abundant, efficient, and spectrally well matched to high-performance silicon solar cells. These rare earth ions are natural four-level system, reasonably transparent to their own radiation, and therefore capable of generating high optical concentrations. Neodymium's and Ytterbium's one disadvantage are their relatively poor overlap with the visible spectrum, meaning that it will require sensitization in the visible spectrum, as Figure 2 shows. It has been shown that transition metals can efficiently transfer energy to these rare earth ions, which will provide the broad sensitization in the visible and near infrared region required. Finally, these ions can be combined with high throughput and chemically robust glass making process for a low cost and stable LSC. Figure 1 shows a schematic of the system. Sensitized Neodymium and Ytterbium should enable single LSC matched to silicon with efficiencies exceeding 10%.
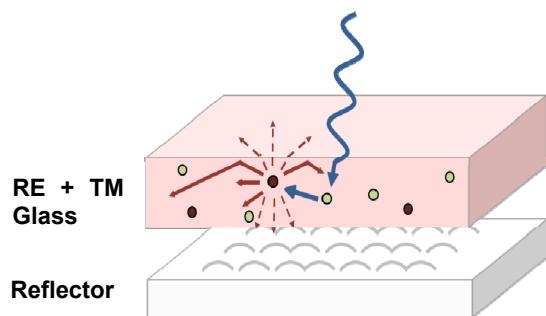

Figure 1: A schematic representation of an LSC. The LSC consists of transition metal (TM) and Nd^{3+} doped glass and a diffuse back reflector. Solar radiation incident on the LSC is absorbed by the Nd^{3+} and reemitted as infrared radiation. The solar spectrum not absorbed by Nd^{3+} is captured and transfered by the transition metal and through nonradiative energy transfer to Nd^{3+}.
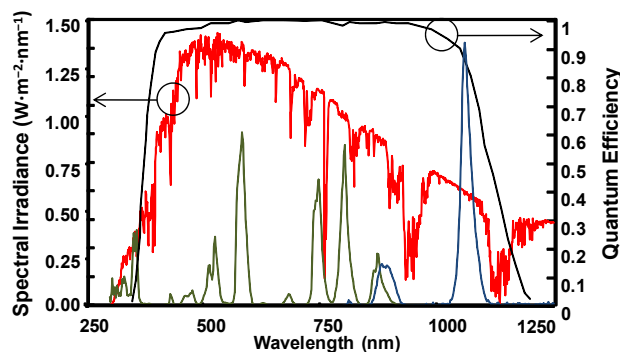
Figure 2: A comparison between the emission and absorption spectra (a.u.) of Nd^{3+} (green and blue, respectively); the AM1.5 solar radiation spectrum (red); and the external quantum efficiency of a Sunpower photovoltaic cell (black) [3].

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6. Non-resonantly Pumped High Quality Factor Lasers

Sponsor

Project Staff
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Lasing Solar Concentrators are expected to have record efficiency in converting light to electricity. They are basically solar pumped lasers, which, due to the low intensity of the solar flux, need to possess a high quality factor (high Q). High-Q lasers are also a key feature in on-chip optical computing. Currently, high-Q lasers can only be resonantly pumped by a second high-Q laser, resulting oscillations in the lasing output [1]. In this work, we show a new pumping technique for high-Q lasers that is independent of the coherence of the pump source. This technique is based on cascaded energy transfer that resembles adiabatic transfer from incoherent to coherent light. Our work opens the way to solar pumped lasers and solves a major bottleneck towards the realization of optical computing.

In the last few months, we have optimized the cascaded energy transfer to yield record Q factor for organic lasers (Q=10000). The energy transfer starts with AlQ_3 molecules that absorb the pump UV light, and transfer energy to DCJTB molecules, which then transfer the energy to Terrylene, which in turn lases at about 680 nm wavelength. The concentration of the Terrylene is about 0.001%. Lasing at such low concentration, (and thus highly transparent media), enables the propagation of the lasing photons for a long distances in the cavity, which is reflected at the high Q factor that we observed. Figure 1 show typical power spectrum measurement, with FWHM at measurement limit of 0.07 nm.

The observed Q factor is : $Q = \vartheta\tau = \frac{\vartheta}{\Delta\vartheta} = \frac{\lambda}{\Delta\lambda} \sim \frac{700}{0.07} = 10000$ and is limited by the spectrum analyzer measurement limit. Our current goal is to measure the real Q-factor at this optimized configuration by measuring the life-time of the photon in the cavity. For this measurement we use a streak camera. This allows us to differentiate between DCJTB and Terrylene emissions. We examine whether the Q factor of the Terrylene is significantly longer than the one of the DCJTB. Also we the measure Q factor dependence with the concentration (absorption) of Terrylene. Preliminary measurements of photon life time vs concentration of Terrylene are shown in figure 2. These results suggest Q factor greater than 500000.

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