

owing to the fact that distinguishing such closely spaced optical frequencies with passive optical filters is difficult. In this respect, the Raman scattering process has an advantage in that the 15.6 THz shift in silicon corresponds to a broader wavelength shift from, for example, 1.55  $\mu\text{m}$  to 1.686  $\mu\text{m}$ . With further work, perhaps the SBS frequency shifts can be increased. Frequency shifts of 50 GHz would enable more practical passive filtering of the optical signals involved in SBS in a number of platforms.

With this foundational result, it has become possible to engineer efficient SBS in nanoscale silicon waveguides with high enough gain to see use in various applications (see Table 1 for a summary of characteristics and comparison to related works in various systems). For example, we may soon see compact chip-scale Brillouin lasers in this material platform.

Owing to the integrated nature of the device and its relatively compact footprint, this approach could eventually be used in large-scale integrated optical systems. What is also exciting about this result is that it has been realized in silicon-on-insulator, one of the world's most commercially accessible material platforms, and without the use of exotic materials or processing steps. It is thus possible that devices similar to these could one day be constructed in complementary metal-oxide-semiconductor (CMOS)-compatible commercial facilities. The work by Kittlaus *et al.* may be a first step towards the utilization of Brillouin processes as an additional tool for on-chip optical nonlinearities and non-reciprocal light flow. □

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## ORGANIC OPTOELECTRONICS

# Stable multilevel memories

Combining a photochromic molecule with a semiconducting polymer yields an organic non-volatile, multilevel memory with a current output that can be switched and controlled by light.

Maryam Ebrahimi and Federico Rosei

The fields of organic electronics and optoelectronics<sup>1</sup> exploit thin films made of electroactive and/or photoactive molecular materials to perform various functions. These tasks can include light emission as in the case of organic light-emitting diodes (OLEDs)<sup>2</sup>, the conversion of sunlight to electrical energy as in organic photovoltaics (OPVs)<sup>3,4</sup>, field-effect current modulation as in organic field-effect transistors (OFETs)<sup>5</sup>, and current modulation and light emission in organic light-emitting transistors (OLETs)<sup>6</sup>. Now, writing in *Nature Nanotechnology*, Leydecker *et al.*<sup>7</sup> add to this arsenal with the demonstration of a stable multilevel organic memory device (OMD) that is controlled by light. Once optimized, such an approach could become an alternative to conventional data-storage technologies based on silicon.

The advantages of using organic materials include the promise of low-cost solution processing and the potential for plastic substrate devices that are ultrathin, large area, lightweight, multifunctional and mechanically flexible. Organic optoelectronics has already been successfully integrated in several commercial

applications that have become part of our daily lives (for example, OLED-based flat-panel displays). However, the development of robust, high-density OMDs<sup>8,9</sup> is still ongoing.

The purpose of a memory device is to store and access binary digital data in a compact physical element that is coupled to a central processing unit<sup>10</sup>. Electronic memories can be volatile (for example random-access memories, in which the information is lost unless constant power is applied) or non-volatile (for example read-only memories (CDs and DVDs) and flash memories). The key performance parameters in memory devices are cost, speed of data storage and/or transfer, size, power consumption, the device lifetime in terms of the number of rewrite cycles, cycling stability and data retention time. Writing and reading data bits using different physical processes such as light and charge transport can improve device reliability.

For a few decades, scientists have attempted to fabricate stable memories that use light inputs to store information in bistable photochromic materials (which undergo a light-activated reversible change

between two states). Leydecker *et al.*<sup>7</sup> have now demonstrated that a marriage of photochromic molecules with organic semiconductors is a potentially winning approach to realize robust, high-density, non-volatile, multilevel organic memories that exhibit high switching ratio, excellent data retention time, high field-effect mobility and low programming voltage.

The underlying idea is to tune the charge transport (and thus flow of electrical current) in a composite organic semiconductor by combining the photoswitching capability of a photochromic molecule with the electroactive properties of another molecule or polymer.

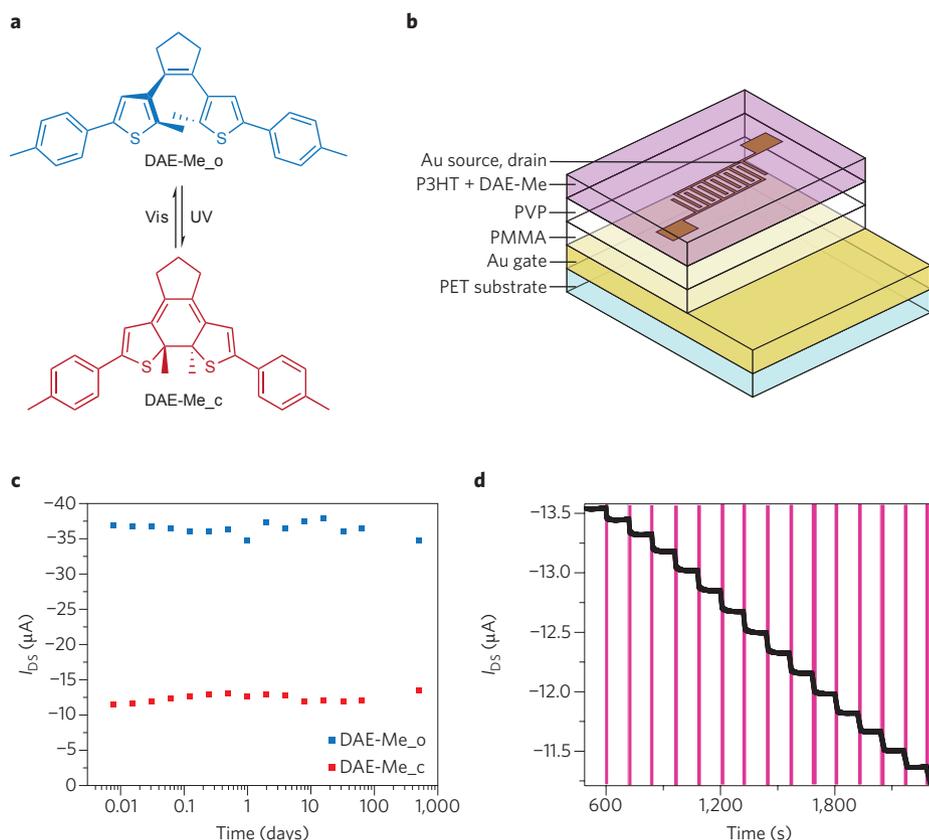
Several materials and device structures with bistable states (a requisite for an OMD) have already been demonstrated<sup>9</sup>. In previous work, an ingenious approach to create phototunable energy levels, thereby forming bistable states, by blending diarylethene (DAE, a small photochromic organic molecule) with the well-known organic semiconducting polymer poly(3-hexylthiophene) (P3HT) was reported<sup>11</sup>. The photochromic molecule (DAE or its

derivatives) is used to tune charge transport in the organic film, by keeping the highest occupied molecular orbital (HOMO) energy level of the open form state of DAE inaccessible to holes. DAE derivatives are an attractive option for this application as they offer high fatigue resistance, thermal stability for both switching states and states with different electronic properties. The photomodulation and performance of an organic transistor can be optimized by the choice of which DAE derivatives and which polymer (for example, P3HT) or small-molecule semiconducting matrix is used in terms of their chemical compatibility and reciprocal energetics<sup>7,11,12</sup>.

Memory devices can be realized from transistors, capacitors and resistors. In their elegant approach, Leydecker *et al.* blend DAE-methyl (DAE-Me, which exhibits excellent photomodulation) and P3HT to realize a composite organic film that acts as an active layer in a transistor (three-terminal) device (Fig. 1b). Transistors are able to implement a signal addressing function in two-dimensional memory arrays that would not be possible if using diode-only circuits. The three-terminal architecture makes it possible to tune the rate of charge carriers via the intensity and duration of a specific wavelength of illumination: monochromatic light at  $\lambda = 313$  nm (write) or 546 nm (erase) enables switching between the open (on) and closed (off) states (Fig. 1a). The insertion of DAE-Me within a P3HT matrix thus allows light to control the output source–drain current,  $I_{DS}$ , in a three-terminal organic transistor. DAE-Me<sub>o</sub> (open) and DAE-Me<sub>c</sub> (closed) states act as scattering centres for charge carriers and hole acceptors, respectively.

The authors showed that their memory devices can be fabricated on two types of substrate: either octadecyltrichlorosilane (OTS)/SiO<sub>2</sub> or flexible polymeric sheets of polyethylene terephthalate (PET). In the former case, chemisorbed self-assembled monolayers of OTS were formed on SiO<sub>2</sub>, with the aim to create a smooth and uniform interface for efficient charge transport, which enhances the device's electrical performance. The organic transistor exhibited optimal performance (in terms of mobility and switching ratio) for a blend with 20% weight DAE-Me, with stable performance over 70 write–erase cycles. The data retention capability exceeded 500 days (Fig. 1c) with excellent read-out accuracy evaluated through current measurements in the dark upon switching cycles, and the multilevel memory attained data storage over 8 bit (256 levels).

For devices fabricated on the flexible PET substrate (Fig. 1b), due to the higher



**Figure 1** | Principle and operation of an organic memory unit. **a**, Chemical formulae of the employed DAE in its open (on; top) and closed (off; bottom) forms. **b**, Structure of the memory on a PET substrate. PMMA, poly(methyl methacrylate); PVP, poly(4-vinylphenol). **c**, Measurement of  $I_{DS}$  (for a memory on a SiO<sub>2</sub> substrate) after different times of storage in the dark at fixed drain–source voltage  $V_{DS} = -10$  V and gate–source voltage  $V_{GS} = -60$  V to illustrate the retention capability of the memory in the open and the closed states. **d**,  $I_{DS}$ –time curve showing how a series of changes in illumination light at 313 nm can create a multilevel memory (5 to 20 s incremental irradiation steps, pink lines). Figure reproduced from ref. 7, Nature Publishing Group.

roughness and thickness of the film, the switching between open and closed states occurred at a slower pace than when using a SiO<sub>2</sub> substrate. The device exhibited a field-effect mobility comparable to that obtained on SiO<sub>2</sub> (over  $10^{-3}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) with the dynamic  $I_{DS}$ –time measurements (Fig. 1d) indicating the ability to work as a 4-bit multilevel memory (16 distinct levels). In addition, the electrical characterization of the device in both closed and open forms revealed that it was very stable, with no significant current variations after 1,000 bending cycles. However, the minor loss of carrier mobility and shift in threshold voltage leave room for improvement.

Such organic three-terminal memory devices fabricated on flexible substrates by Leydecker *et al.* could ultimately be useful for future applications in wearable electronics and optoelectronics that are inaccessible to silicon-based technology. They may also prove useful for applications

involving organic sensors, biosensors, energy storage and solar energy conversion.

Future work should focus on molecular-level studies of the interaction between the components of the organic blend (different photochromic molecules combined with other organic semiconductors) and the role of the solvent. The effect of the injected charge on the photochromic molecule's isomerization yield may depend on the type of polymer, and this aspect should be studied in more detail. In addition, P3HT is known to be sensitive to air and should therefore ideally be replaced by an alternative air-stable p-type semiconductor with similar energy levels, leading to longer-term device performance. The judicious choice of the photochromic–electroactive pair and the ability to make a uniform blend (that is, with a high degree of intercalation of the photochromic units within the polymer matrix) should be primary considerations in future

technological developments of OMDs based on this concept. Once these challenges are addressed, the main remaining issue will be fast-prototyping over large areas. In summary, high-density non-volatile flexible OMDs for data-storage applications now look to be within reach. □

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## X-RAYS

# Strong coupling of light goes nuclear

Evidence of strongly enhanced coupling of light with nuclei offers the potential to extend applied quantum optics to the hard X-ray regime.

Brendan Dromey

Applying the principles of quantum optics to functional systems relies on the coherent interaction of large numbers of photons with extended ensembles of atoms. Over the last half century, the ability to precisely control these interactions has taken the design of such systems down to the nanoscale<sup>1</sup>. This research has been the driving force behind several technologies from high-specification lasers for science and manufacturing, to novel imaging techniques for biomedical applications and the development of photonic devices for the optical telecommunications and semiconductor industries. To date, however, applications in quantum optics have largely been based around the interaction of optical light with electrons in the outer shells of atoms (or molecular/chemical bonds). This is because at these wavelengths there are numerous resonant transitions that permit strong coupling between the incident electromagnetic radiation and the target atoms. Unfortunately, extending this control to shorter wavelengths becomes increasingly difficult as the likelihood for resonant interactions to dominate over non-resonant processes diminishes rapidly, especially in the hard X-ray regime.

Writing in *Nature Photonics*, Johann Haber *et al.* demonstrate experimentally that it may be possible to extend quantum optics research into the X-ray region of the spectrum by coherently controlling the interaction of light with quantum states of the nucleus<sup>2</sup>. In their work, Haber and colleagues from the Deutsches Elektronen-Synchrotron (DESY), Helmholtz-Institut Jena and the European

Synchrotron Radiation Facility (ESRF) provide the first unambiguous evidence that collective effects arising from the interaction of light (here synchrotron radiation at X-ray wavelengths) with nuclei in a carefully constructed stack of isotopic bilayers can be enhanced and tuned to a degree where potential applications can, in principle at least, be investigated. This in turn provides a significant breakthrough for the potential of applications arising from the burgeoning field of nuclear quantum optics.

To put it simply, nuclear quantum optics takes the principles of quantum optics for the interaction of visible light with the electronic shells of atoms and applies them to the quantum states of the nucleus. The key to the long-term success for this field will be to achieve quantum physical control of X-rays and their interactions with nuclei so that the processes under investigation can happen collectively, or in a coherent manner. If this can be achieved the potential benefits for researchers studying the microcosm are promising. It will allow the technological breakthroughs provided by optical quantum systems to be transferred directly to the short-wavelength region of the spectrum. This may enable developing narrowband X-ray and  $\gamma$ -ray lasers based on resonant nuclear transitions<sup>3</sup> as well as novel techniques for directly studying the structure and dynamics of nuclei embedded in matter.

Over the last few decades, research into nuclear quantum optics has focused largely on the nuclear forward scattering of X-ray synchrotron radiation from Mössbauer nuclei in a crystal lattice. These nuclei are unique in that once embedded in a lattice array they do not exhibit a redshift

(or energy loss) in the emission of an X-ray/ $\gamma$ -ray photon due to nuclear recoil. This property implies that interactions can be finely tuned with the addition of magnetic fields (either externally applied or imposed through interactions with surrounding atoms in the lattice) to study nuclear dynamics and hyperfine structure directly<sup>4</sup>, demonstrating the possibility for coherent control of the quantum states of the nucleus<sup>5,6</sup>.

More recently, researchers have managed to improve the signal observed from nuclear forward scattering by the careful assembly of Mössbauer nuclei in planar cavity waveguides<sup>7,8</sup>. This has improved the collective light–matter interaction through the excitation of cooperative radiative eigenstates of the nuclei. Importantly, it also allows for the selective placement of <sup>57</sup>Fe nuclei in the nodes and antinodes of the propagation mode in the cavity. This has facilitated the observation of the collective Lamb shift<sup>9</sup> and electromagnetically induced transparency<sup>10</sup> at X-ray wavelengths. However, while ground breaking, this method still suffers from the problem that the overall interaction is only weakly enhanced. This limits its use for ambitious applications based around a strong coupling of hard X-rays with matter<sup>3</sup>.

To address this problem, Haber *et al.* draw on the principles of photonic bandgap materials to enhance light coupling with the nuclei of selected atoms. Photonic crystals, as they are also called, have proven to be extremely successful at manipulating light at optical wavelengths through periodic modulation of the refractive index (or dielectric constant) within the