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Lasing from exciton-polariton coupling in metal organic frameworks

Research Objectives

The research group studies ultrastrong coupling and investigation on double-threshold lasing behaviour in metal-organic framework (MOF) microplates.

Detail

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Bio

Professor Ji Wei's research interests cover nonlinear optics and ultrafast nonlinear spectroscopy, for nonlinear optical and quantum materials which have photonic and optoelectronic applications. He has co-authored over 230 research papers, and his h-index is above 50. Recently, his research interests have been extended to optoelectronics with 2D materials and metal-organic frameworks.

Dr Dileep Kottlil's research interests include nonlinear optics, nanomaterials, strong light-matter interaction in metal-organic frameworks, etc. He received his BSc in Physics from MES College Ponnani, India (2011). He completed his MSc Physics (2013) from IIT Madras, India. He completed his joint PhD from National University of Singapore, Singapore and IIT Madras, India. Currently, he is a post-doctoral fellow at National University of Singapore.

Funding

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Collaborators

- Prof C Vijayan, Indian Institute of Technology Madras, India
- Prof Parimal K Bharadwaj, Indian Institute of Technology Kanpur, India
- Dr Mayank Gupta, National University of Singapore, Singapore

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Personal Response

How customisable do you think is this new method for cavity fabrication?

1. In our work, we used rhodamine B as the dye molecule. Replacing this molecule with a wide variety of other available dye molecules can potentially realise laser cavities emitting almost all the wavelengths in the visible range. However, chemical synthesis to realise such stable crystals is a big challenge.
2. Our MOF crystal is a stack of many 2D layers. Similar measurements and studies on 1D and 3D MOFs could have an impact on the efficiency and threshold of resulting lasers.

Lasing from exciton-polariton coupling in metal-organic frameworks

When a molecule is trapped inside a cavity, sometimes strange things happen – including some unusual physics. By adjusting the cavity size to match some of the unique properties of the molecular species trapped inside, it is possible to create special hybrid states that can modify the optical properties of a given system. Professor Ji Wei and Dr Dileep Kottlil at the National University of Singapore have been designing new systems that can make use of exciton-polariton couplings to be used as lasers.

Light is a strange thing in many ways. A central concept in quantum mechanics, for example, is that light can behave as both a particle and a wave. Light can also have unusual interactions with systems in confined optical cavities. A high-quality optical cavity usually consists of two reflective surfaces, one at each end, so once the light enters the cavity, it will bounce many times between the two ends of the cavity, back and forth. The higher the quality, the smaller the amount of light lost on each trip.

Molecules and materials each have their own optical properties, including what colours of light they absorb. When designing new molecules for optical applications, scientists often try to tune these optical properties to enhance them

for the given purpose – for example, designing molecules that emit light more efficiently for use in television displays.

HOW IT'S DONE

The most common way of trying to tune the optical properties of molecules is to change the chemical composition of the molecule. Certain elements can change how the electrons are distributed in a molecule and, therefore, how it interacts with light. However, this is not the only method available for altering the optical properties. Another approach is to create an optical cavity and embed the molecule into it.

If the optical cavity is of sufficiently high quality, a so-called 'strong field interaction' occurs between the molecule

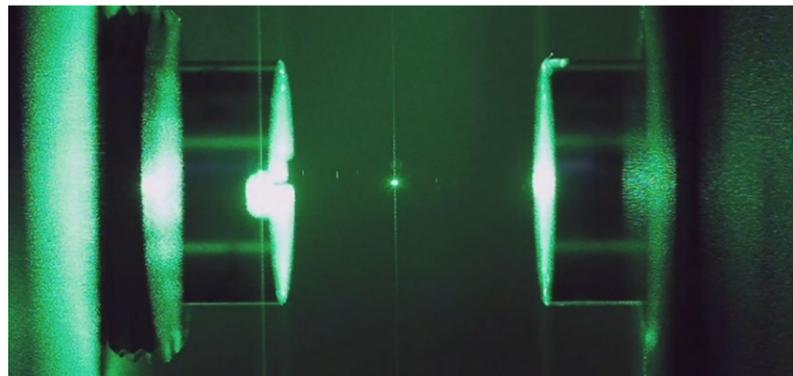
and the cavity. This causes the creation of new absorption lines of the molecules at higher and lower energies than its original unperturbed absorption. These new states are known as polaritonic states, and they arise as a result of this strong coupling.

Professor Ji Wei and Dr Dileep Kottlil at the National University of Singapore are researching new ways to create polaritonic states without even having high-quality optical cavities. Rather than being made of two mirrors, these cavities are made of special materials known as metal-organic frameworks (MOFs). Engineering cavities to the conditions required to observe strong field coupling effects is no easy task; using MOFs is a cost-effective way of making new cavities that could aid the design of new materials to be used as part of laser systems.

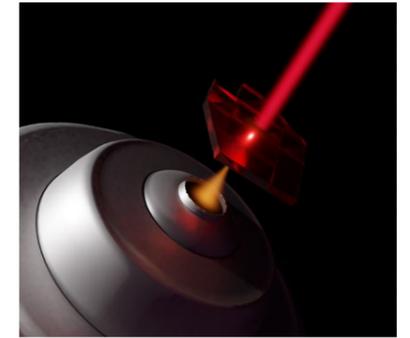
METAL-ORGANIC FRAMEWORKS

MOFs are a type of material made up of a combination of metal ions coordinated to organic linkers. The linkers bind the whole framework together to keep it stable, and the metal ions are arranged in shapes to form one-, two-, or three-dimensional structures. Often, these materials have long pores and channels running through them, similar to porous pieces of rock, and small molecules can pass through these channels and interact with specific sites.

It is possible to create MOFs with a variety of different structures and control the pore sizes and chemical composition at different points in the structure. This means that MOFs can be created with pores small enough to block certain chemical species from ever entering, or only bind particular chemical substrates of



Glass nanoparticle suspended in an optical cavity.



Wei and Kottlil have designed MOFs that can be easily excited with multiphoton processes, to be used as lasers.

interest. MOFs are therefore an incredibly interesting platform for performing controlled chemical reactions and have been of great interest as a material for trapping carbon dioxide.

Wei and Kottlil are using MOFs in a very different way. They have been taking advantage of their excellent chemical binding capabilities to bind a dye molecule, rhodamine B, into the MOF structure. They have then been using MOFs not for controlled chemistry, but as a cavity to modify the optical properties of the dye.

One of the key advantages the researchers found is that this method of creating a polariton cavity is incredibly straightforward and cost-effective compared to traditional approaches, which often involve trying to coat layers of the reflective material and chemical substrate to build up the cavity. One of the big challenges of building up individual layers to produce cavities is that, for many chemical substrates, it is very challenging to keep all the layers of even thickness. This means that the cavity is not uniform in thickness, and the optical properties of the trapped molecules vary at different positions throughout the cavity.

MAKING A LASER

Wei and Kottlil were interested in trapping dye molecules in cavities for their potential to be used as a laser medium. Dyes appear brightly coloured because they absorb light very strongly, but they can emit light very efficiently too. Lasers

work by containing a lasing medium which absorbs light of a given colour to trigger a stimulated emission process and emit light of a colour of longer wavelength. By using substrates with particular optical properties, it is possible to trigger an optical-gain process which will lead to the desired lasing.

As the light intensities used to excite a lasing medium can be very high, laser media need to be highly photostable. To make the lasing process sufficiently efficient, the medium also needs to be an efficient emitter at the right wavelength.

However, the research team discovered that their MOF laser was not just efficient at absorbing a single photon at a time to trigger lasing, but that it could also be used to absorb multiple photons. Sometimes, to trigger emission from a molecule, either a single, more energetic

There are a growing number of medical applications where light is used as a treatment in itself to kill cells or bacteria, or to activate a photosensitive medicine.

photon can be absorbed, or the same effect can be achieved through multiple, less energetic photons. The problem with multiphoton excitation processes is that, for most molecules, they are very statistically unlikely compared to events where a single photon is absorbed. Professor Ji's recent work on this topic has been highlighted as the back-cover art in the journal *Advanced Functional Materials*, Volume 30, Issue 32.

THE FUTURE OF MATERIALS?

There are some good reasons why

scientists like Wei and Kottlil want to create materials that absorb multiple photons efficiently. There are a growing number of medical applications where light is used as a treatment in itself to kill cells or bacteria, or to activate a photosensitive medicine. Our skin and tissues absorb most visible photons, but as the photons become longer in wavelength, towards the infra-red, they can penetrate further past the skin. This is known as the 'biological window' – a measure of what colours of light can pass through our skin to be potentially used for treatment.

The MOFs designed by Wei and Kottlil could easily be excited with multiphoton processes. They could open the way to interesting medical applications and microscopy work, where multiphoton excitation can be used to take less noisy microscope images.

Wei and Kottlil are hoping their new fabrication method will open up a cheap, straightforward, and reproducible way of creating

cavity systems to explore strong field phenomena. One of the greatest challenges in this area of optics research is the fabrication methods, so simplifying these will mean a playground of new systems that can be used to explore the fundamental physics involved in the exciton-polariton couplings in these systems. As optical cavities can also be used to tune the chemistry of molecular species, there are a number of exciting new applications that the ability to make new cavity systems could realise.



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