Ultrafast manipulation of photon transport and molecular beams

Contact *o.muskens@soton.ac.uk*

T. Strudley, P. Venn, H. Ulbricht, O.L. Muskens

School of Physics and Astronomy, University of Southampton
Southampton, SO17 1BJ, UK

Introduction

A femtosecond Ti:Sapphire laser was used to investigate the transport of light in random nanomaterials and the manipulation of cold molecular beams. Both projects have yielded successes opening avenues into the ultrafast laser manipulation of matter.

Transport of light in random nanomaterials

The existence of a speckle pattern from light propagating through a random medium is due to the coherent interference of the many different propagation paths available. Recently there has been considerable interest in exploiting these coherent interference effects for applications such as focusing of light through opaque media[1]. Random media also present the opportunity to investigate fundamental mesoscopic transport properties of waves such as localisation.

The first project aimed to achieve ultrafast control of the mesoscopic transmission modes through a random array of gallium phosphate nanowires. Mesoscopic transport arises when the probability of light paths crossing inside the medium becomes finite. For three dimensional materials these effects canonly become pronounced in very strongly scattering materials in which the mean free path is smaller than the optical wavelength. The strong scattering properties[2] of the nanowires, coupled with negligible absorption of light at wavelengths above the band gap, makes them a promising subject for the investigation of mesoscopic transport effects.

We have recently demonstrated a new phenomenon in which the coherent light paths in random scattering can be sufficiently modulated by an ultrafast light pulse to achieve a dephasing of the transmission speckle[3]. Following on from this, we have studied the effects of ultrafast dephasing in the mesoscopic transport regime.

Experimental Method

A schematic of the experimental setup is presented in Fig 1. A transmission microscope consisting to two high numerical-aperture microscope objectives was used. 800 nm light was focused on to a sub-micrometre spot on the sample by a 0.9 NA objective, and the transmitted light was collected by a 1.3 NA oil-immersion objective and imaged onto a CMOS camera. The sample was mounted on a translation stage to probe many different positions on the sample in order to build up a statistical picture of the mode intensity distribution. Part of the laser output was frequency doubled and used as a pump for excitation of the nanowires following the method of Ref. [3].

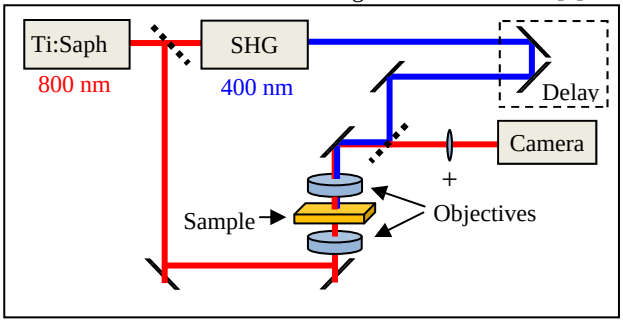

Fig 1: Schematic of experimental setup

Results

A plot of the total transmitted intensity normalised to the ensemble average, s_a, for different positions on the sample is shown in figure 2. After all sources of random fluctuations in our setup were corrected for, significant intensity fluctuations can still be seen, characteristic of mesoscopic transport.

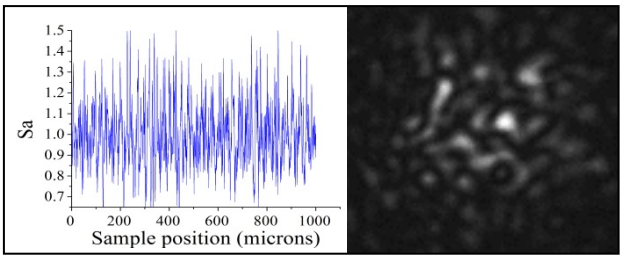

Fig 2: (left) Total transmitted intensity fluctuations with position, (right) typical speckle image.

Figure 3 shows a histogram of the fluctuations in the angular intensity, s_{ab}, of each speckle, normalised to the average speckle. Without mesoscopic correlations, the distribution is expected to follow a negative exponential law known as Rayleigh statistics (blue line). Fluctuations in s_{ab} result in the tail seen in the distribution of the data points (circles). This is again characteristic of mesoscopic effects and can be fitted using an analytical formula obtained from random matrix theory[4] (red line). Good agreement is found with a fit for 25 independent transmission modes.

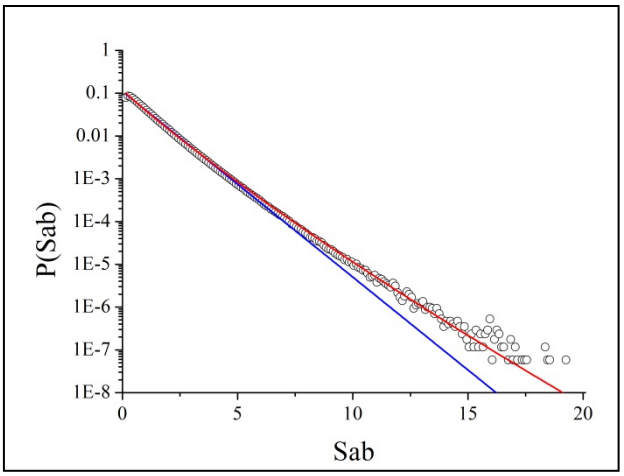

Fig 3: Distribution of angular intensities normalised to ensemble average (Sab).

As a next step, we performed pump-probe measurements in order to observe changes in the transmission statistics due to dephasing. Results will be presented in future work.

Conclusion

Our studies of light transport through random nanowire arrays have allowed direct assessment of mesoscopic fluctuations in the transmitted light intensity. This is an excellent starting point to investigate the effects of ultrafast dephasing on the mesoscopic transport of light in complex photonic media.

Manipulating the motion of large neutral molecules with the off resonant dipole force

When manipulating the motion of particles there are typically two experimental regimes: resonant interactions for atomic systems[5], and optical tweezing[6] for large particles of several microns. The ability to manipulate molecules is still in its infancy; recently resonances in a molecule (SrF) have been exploited for the first time[7]. It is not however possible to scale this interaction for larger molecules as the resonance is inherent to the molecule. Instead alternatively techniques are required to control larger molecules.

It was proposed by Seidman[8] that an off-resonant laser could be used to induce a dipole in a molecule to alter the centre of mass motion and create a molecular lens. The presence of an off-resonant field creates a potential $\boldsymbol{U = -\frac{1}{4}\alpha E^2(x,y,z,t)}$, where $\boldsymbol{\alpha}$ is the static polarizability of the molecule and $\boldsymbol{E(x,y,z,t)}$ is a time varying electric field. Using a Gaussian shaped laser, a dispersive gradient force, $\boldsymbol{F = -\nabla U}$, is experienced by a free molecular beam allowing the creation of a molecular lens. This acts in the same way a conventional lens, but instead of using matter to focus light, light is used to focus matter.

Molecular lensing of small molecules (CS_2, Benzene) has previously been observed using pulsed nanosecond off-resonant lasers[9,10]. The aim of our experiment was to realise a molecular lens for larger molecules such as tetraphenylporphyrin (TPP), which has a mass of 615amu and polarizability of $\boldsymbol{105 Å^3}$. The lens was used to increase the number of molecules in a free beam by improving the beam collimation. Simulations predicted that by using the large peak powers of a femto-second laser an increase of up to 20% in the number of TPP molecules in a molecular beam reaching a detector with diameter 1mm. This increases the luminosity of our molecular beam and is a general system that could be used to improve any molecular beam source as long as the interaction is off-resonant and below the ionization threshold of the molecule. Here we present our experimental setup and results in the optical manipulation of a thermal beam of TPP.

Experimental Method

The experiment consisted of two interconnected vacuum chambers with typical pressures 10^{-6} and 10^{-7} mbar for the source and detection chambers respectively. In the source chamber an oven sublimates molecules at typical temperatures of 800K, creating a vertical beam source. The molecules then pass through a helical velocity selector and a 1mm collimation hole to give a typical beam collimation of 1mrad**.** In the detection chamber the molecule beam passes through the focussed laser, with the wavelength chosen so that it was off resonant for TPP (800nm). This creates the potential needed for lensing, and the molecular beam then travels 0.6m to a quadrupole mass spectrometer (QMS) where the number of molecules detected was measured when the laser was on or off. A neutral density filter was used so that the power of the laser could be varied. Due to the high number of dark counts (triangles in Fig.4) associated with the detection of large molecules measured were averaged over 50 seconds. A schematic of the experiment is given in Fig 3.

Results

Fig 4 shows the results from an experimental run where an increase of 10.0±1.0% in average molecule counts was observed for a thermal beam of TPP. With the used laser parameters we do not expect ionization or absorption of photons to have had a significant effect on the results, as ionisation of the molecules at the position of the lensing laser would have created a decrease in detected molecules. In addition the estimated number of absorbed photons per molecule and laser pulse is much smaller than 1.

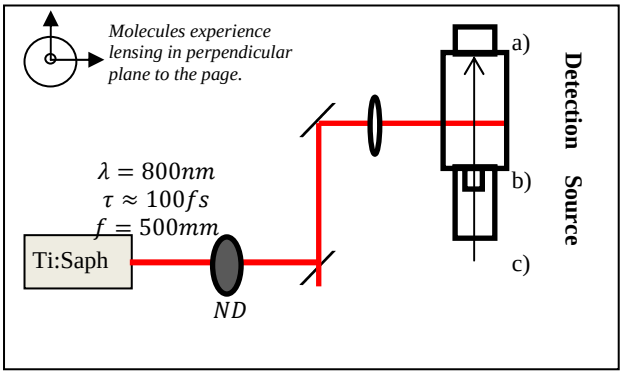

Fig3: Experimental setup of molecular lensing experiment, a) is a QMS molecule detector, b) is a helical velocity selector $\frac{\Delta v}{v} = 4.4\%$ and c) is the sublimation oven, T=800K. The arrow shows the direction of the molecule beam.

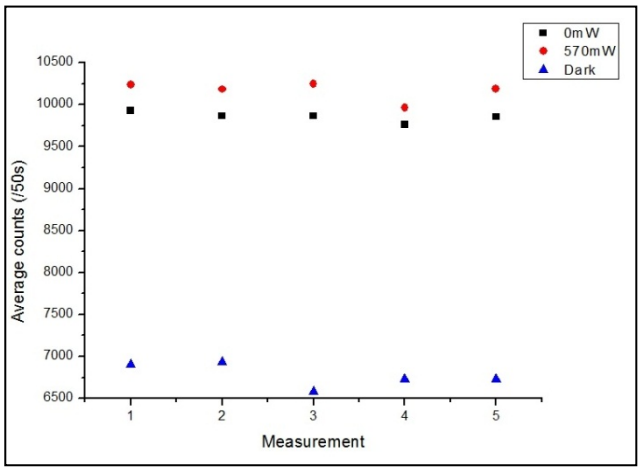

Fig 4: Average molecule counts for a free molecular beam of TPP interacting with the focussed femtosecond laser. The dark counts from the detector are indicate by the triangles, the circles demonstrate the number of molecules detected when the laser has an average power of 570mW and the squares indicate a measurement of count with no laser present.

Conclusion

We have demonstrated that by using the off-resonant dipole interaction a molecular lens can be created that collimated a free molecular beam leading to a 10% increase in molecular flux. We think that our scheme represents an important step in the development and realisation of generic schemes to optically manipulate and cool large molecules.

Acknowledgements

The use of the UFL1 from the Laser Loan Pool is gratefully acknowledged by the authors. We would like to thank Peter Horak for assistance with the modelling program for the optical manipulation. Funding agencies: EPRSC, SEPNET, and FQXi.

References

[1]. G. Lerosey et al., Science 315, 1120 (2007)
[2]. O. L. Muskens et al., Nano Lett. 9, 930 (2009)
[3]. M. Abb, E.P.A.M. Bakkers, O. L. Muskens, Phys. Rev. Lett. 106, 143902 (2011)
[4]. Th. M. Nieuwenhuizen, M. Van Rossum, Phys. Rev. Lett. 74, 2674 (1995)
[5]. A. Ashkin, Phys. Rev. Lett., 25, 1321-1324 (1970)
[6]. A. Ashkin et al., Opt. Lett. 11,5 (1986)
[7]. E.S Shuman et al., arxiv :0.909..2600v1 (2009)
[8]. T. Seidman, J. Chem. Phys. 106, 7 (1996)
[9]. H. Stapelfeldt et al., Phys. Rev. Lett. 79, 15 (1997)
[10]. Chung et al., Journ. Chem. Phys., 114, 19 (2001)