Cooling materials using interaction with light has come a long way in the past 20 years. Researchers at the recent 2013 Photonics West showed that they can cool new types of materials and more can be expected in the future using new approaches.

David Pile

At the beginning of each year, San Francisco hosts one of the biggest and broadest optics conferences in the world. This year, SPIE’s Photonics West conference took place on 2–7 February 2013 at the Moscone Center, which is conveniently located close to the Embarcadero, Mission District and numerous other popular destinations.

As the weight of the 415-page technical program indicates, the meeting was incredibly broad. Major themes were grouped under ‘sub-conferences’ named ”Green Photonics”, ”BOS”, ”LASE”, ”MOEMS-MEMS” and ”OPTO”. The laser cooling of solids and dense gases (also known as optical refrigeration), although not one of the ‘biggest’ topics at the meeting, provided some focused sessions that demonstrated the significant recent progress. Optical refrigeration is of interest because it can be realized using compact, cryogen-free systems and does not generate vibrations.

One of the early sessions addressed the problem of cooling optically important rare-earth-doped material systems. Seth Melgaard and colleagues from the USA (Air Force Research Laboratory and the University of New Mexico) and Italy (Università di Pisa) discussed several milestones. They have cooled a few types of particles using laser-generated plasmas.

Wim Leemans of Lawrence Berkeley National Laboratory gave a plenary talk on the acceleration of particles using laser-generated plasmas. Melgaard explained that they improved the cooling efficiency by investigating the effect of doping concentration. The ratio of the background absorption to the resonant absorption of the 10%-wt Yb:YLF crystal was reduced giving the 10%-wt Yb:YLF crystal a minimum achievable temperature of 93 K. So far, they have achieved a temperature of 114 K; Melgaard emphasizes that, although this is above the minimum achievable temperature, it is by far the coldest solid-state optical technology in the world. “We have identified a path towards achieving solid-state cooling to the liquid-nitrogen temperature of 77 K,” Melgaard told Nature Photonics. “Because of the trend in parasitic background absorption, elemental analysis was performed and it identified iron as the main contributor to parasitic heating. By reducing the iron concentration through purification of the starting materials, model predictions show that solid-state cooling to liquid-nitrogen temperature is within reach.”

Also on the topic of cooling rare-earth-doped systems, Angel Garcia-Adeva (Universidad del País Vasco) and colleagues from Spain and France discussed work on using light to cool erbium-doped oxysulphide crystal powders. Garcia-Adeva explained to Nature Photonics that lanthanum oxysulphide, a uniaxial P3m wide-bandgap semiconductor material, is an excellent host lattice for trivalent rare-earth ions, as its maximum phonon energy of about 400 cm⁻¹ enhances efficient upconversion processes while strongly suppressing nonradiative multiphonon losses. “Our group is currently investigating Er³⁺-doped La₂O₂S crystal powders as a promising candidate for all-optical cooling. This material exhibits an efficient infrared-visible upconversion under excitation in the 800–870 nm band,” Garcia-Adeva told Nature Photonics. “Indeed, we have obtained efficient upconversion-assisted local cooling when pumping in resonance with a two- or three-phonon annihilation process. Even though this investigation is still underway, our preliminary results suggest that efficient bulk optical cooling could soon be achieved in this system.”

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transitions between the excited and ground states of isolated ions (anti-Stokes fluorescence in rare-earth-doped insulator hosts) or between the conduction and valence bands (laser cooling with direct-bandgap semiconductors). In contrast, the cooling mechanism in a rare-earth-doped direct-bandgap semiconductor system (Yb\(^+\)-:InP) consists of laser excitation of Yb\(^+\) ions followed by thermal quenching of excited ions accompanied by phonon absorption, which provides the cooling; band-to-band radiative recombination in the InP host at the end of the cooling cycle removes energy from the system. Nemova says that, in contrast to cooling with conventional rare-earth-doped insulator hosts, in which the pump wavelength must exceed the mean fluorescence wavelength, their system can be pumped at a wavelength shorter than the mean fluorescence wavelength, thus benefiting from the high absorption cross-section of rare-earth ions. The increase in quantum defects between the absorbed and radiated photons may give a higher cooling efficiency than the laser cooling of rare-earth-doped insulator hosts.

Following the success with cooling rare-earth-doped solids, groups have also pursued the optical refrigeration of conventional semiconductors. Recently, Qihua Xiong’s group from Nanyang Technological University in Singapore has accomplished, for the first time, the net laser cooling of group \(\text{II}–\text{VI}\) semiconductor CdS nanoribbons to a temperature of about 40 K below ambient temperature. Xiong explained to Nature Photonics that coupling between strong excitons and longitudinal optical phonons in group \(\text{II}–\text{VI}\) semiconductors such as CdS can be harnessed to facilitate laser cooling by the annihilation of one or more longitudinal optical phonons, leading to several \(k_B T\) of heat being removed in each cooling cycle. He explained that net cooling in semiconductors had not been achieved previously, despite many experimental and theoretical efforts on group \(\text{II}–\text{V}\) gallium arsenide quantum wells. According to Xiong, GaAs-based semiconductors suffer from a high parasitic background absorption and a poor luminescence extraction efficiency, although anti-Stokes upconversion can be readily achieved. The researchers suggest that group \(\text{II}–\text{VI}\) semiconductors may be more promising than group \(\text{II}–\text{V}\) semiconductors for an all-solid-state optical refrigerator down to sub-liquid-nitrogen temperatures.

Others discussed the applications of semiconductor cooling to light-emitting diodes (LEDs). Rajeev Ram from the Massachusetts Institute of Technology in the USA explained how electroluminescent cooling broadens the application scope of optical refrigeration. “By operating an LED as a heat pump, whereby the electrons transport energy from lattice vibrations to incoherent photons, it is possible to realize LEDs in which nearly all of the energy for photon emission originates as heat,” Ram explained. “From the conventional device perspective, the electrical-to-optical power conversion efficiency seems to be greater than 100%. In this operating range, LEDs become brighter and more efficient as their temperature rises and more heat is pumped into the device.” According to Ram, applications for LED’s operating in this electroluminescent cooling regime include mid-infrared sources for sensing in harsh environments and potentially even general illumination.

Also on the subject of LEDs, Volodymyr Malyutenko from the Institute of Semiconductor Physics (Ukraine) has demonstrated an alternative approach to cooling of a commercial green LED operated in continuous-wave mode and self-heated to 150–200 °C. In his set-up, the LED sits on a cooler consisting of a 15 mm × 15 mm × 4 mm silicon wafer pumped by a 1.09-μm-wavelength diode laser. They achieved 5 K cooling in both the LED and the silicon wafer when the pump was operating. The physical concept is based on a light downconversion process initiated by free carriers generated by the pump. The cooling occurs due to the enhancement of infrared (>3 μm) thermal emission in the silicon when the overall energy of multiple low-energy photons escaping the wafer exceeds the above-bandgap energy of the pumped photon.

There was also a session dedicated to novel approaches for laser cooling. Martin Weitz of Bonn University (Germany) reported an experiment investigating the laser cooling of atomic gases by collisional redistribution of fluorescence. Weitz explained to Nature Photonics that this technique is applicable to ultradense atomic ensembles of rubidium atoms with a noble buffer gas pressure of a few hundred bars. Frequent collisions with noble gas atoms in the dense gas system shift atomic transitions to resonance with a laser beam detuned by a few nanometres to the red of an absorption line, while spontaneous decay occurs close to the unperturbed resonance frequency so that the ensemble is cooled. This new laser cooling technique is suitable for cooling high-density ‘macroscopic’ gas ensembles.

The redistribution laser cooling technique is expected to be applied to molecular gas samples in the future.

In the same session, Tal Carmon of the University of Michigan (USA) discussed Brillouin cooling. Carmon explained that although glass tends to be viewed as a stationary medium that interacts minimally with electromagnetic waves, Brownian fluctuations of atoms in solids scatter and Doppler-shift light while exchanging energy with matter. Such scattering from density variations in the form of acoustic waves is called Brillouin scattering; Raman scattering refers to scattering from fluctuations in charge distribution while the molecule’s centre of mass stays stationary. Carmon explained that to enable cooling through scattering, the system should prefer ‘bluer’ wavelengths (anti-Stokes scattering), which extract energy from the material, rather than ‘redder’ photons, which impart energy to the material. “Thermodynamically, however, the Planck distribution suggests that the red Doppler shift dominates over the blue shift, which is true for bulk materials in which light of all colours is equally allowed,” Carmon told Nature Photonics. “To tilt this balance towards the other side and to reverse the energy transfer direction in Brillouin scattering, we used a microresonator in which the blue scattering from sound is resonantly enhanced while red scattering is off-resonantly discriminated.” Essentially they achieved resonant filtering in the system to ‘prefer’ blue, thus counteracting the Planck-distribution tendency to prefer red. In their set-up, cooling was measured from a reduction in the Brownian scattering when the input optical power was increased, which is opposite to previous experiments on Brillouin scattering in which scattering increases with input power. Carmon pointed out that this Brillouin cooling experiment raises the question of whether resonant colour filtering techniques might also be able to support Raman cooling in solids that are expected to have much higher quantum cooling efficiencies.

Clearly there is a lot of activity in the field of optical refrigeration. Precisely how far cooling can go, and for which materials, are matters of significant debate. However, we can no doubt look forward to seeing further rapid progress at next year’s Photonics West, which will be held in San Francisco on 1–6 February 2014.

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