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Los Alamos
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Laser Cooling of Infrared Sensors

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ABSTRACT

We present an overview of laser cooling of solids. In this all-solid-state approach to refrigeration, heat is removed radiatively when an engineered material is exposed to high power laser light. We report a record amount of net cooling (88 K below ambient) that has been achieved with a sample made from doped fluoride glass. Issues involved in the design of a practical laser cooler are presented. The possibility of laser cooling of semiconductor sensors is discussed.

Keywords: laser cooling, laser refrigeration, optical cooling, optical refrigeration, cryocooler

1. INTRODUCTION

Laser cooling of solids is the process of lowering the temperature of a carefully engineered condensed matter system by illuminating it with laser light. This is sometimes referred to as optical refrigeration. Laser cooling offers many important advantages compared to conventional cryocooler technology: compactness, absence of moving parts or fluids, an all-solid-state design, no vibrations, low mass, no generation of electromagnetic interference, high reliability, and extremely long operational lifetime. Laser cooling is particularly attractive for space-based sensor applications where the above features provide important advantages. Other applications include terrestrial sensing for pollution monitoring and law enforcement, gamma-ray detection, and cooling of superconducting devices and electronics. Laser cooling has the potential of achieving much lower absolute temperatures compared to multi-stage Peltier coolers, which are also brittle and incur significant mechanical stress at large temperature gradients. Laser cooling is attained by radiative transfer of thermal energy – not conductive or convective heat flow.

The first demonstration of the effect was made at Los Alamos National Laboratory in 1995 using Yb-doped glass.[1] We report here a new record for the amount of temperature decrease below ambient ($\Delta T = 88$ K) and the lowest absolute temperature of 210 K. Our group is currently involved in an intensive research and development program to push temperatures much lower. There are no fundamental physical limits that prevent realization of absolute temperatures below those possible with thermoelectric coolers or liquid cryogen technology. In this paper, we briefly review the progress made in cooling of glasses and crystals doped with the rare earth elements Yb and Tm. We discuss the prospects for laser cooling of semiconductors such as GaAs, where realization of extremely low absolute temperatures is expected.

2. THE COOLING CYCLE

The possibility of optically cooling of solids was predicted in 1929.[2] This prediction was based on fundamental thermodynamic arguments and of course, did not rely on the existence of lasers, which would not be invented for another 32 years. Even the availability of laser light did not make the problem trivial. Achieving adequate material purity along with careful preparation, polishing, and strict attention to heat management were essential to realization of net cooling.[1] The principle of laser cooling is depicted schematically in Fig. 1. Laser photons with energy $h\nu$ (h is Planck's constant and ν is the laser frequency) excite electrons from a low energy ground

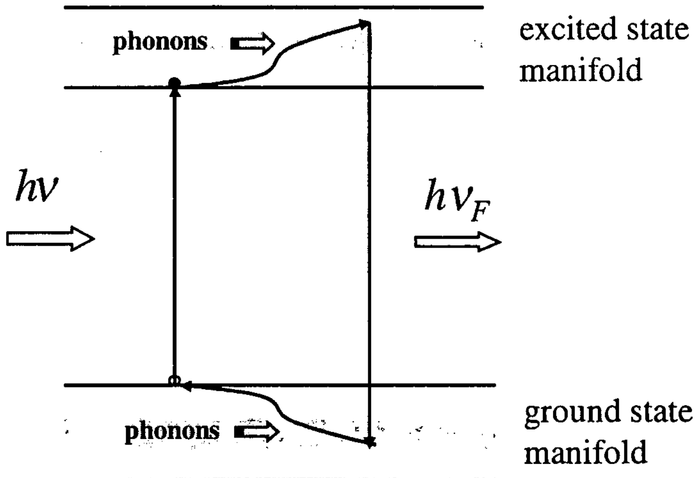

Figure 1. The laser cooling cycle in rare-earth doped crystal or glass. Pump photons of energy $h\nu$ cause excitation from the top of the ground state manifold to the bottom of the upper state manifold. Thermal excitations (phonons) move electrons to higher energy in the upper state where blue-shifted recombination can occur at $h\nu_F$. If this cycle can be made to occur with high efficiency, net cooling occurs.

state to an excited state of the solid. The upper state lifetime – the time excited electrons spend in the upper state before recombining – is long enough that the electrons have a high probability of absorbing thermal energy from the material. The amount of thermal energy gained by the electrons in the excited state is of the order of kT, where k is the Boltzmann constant and T is the temperature of the sample. Relative to the ground state, the energy of the thermally excited electrons is approximately $h\nu + kT$. It is in this state that we wish the electrons to make an optical transition back to the ground state. In doing so, they emit a photon of energy $h\nu + kT$, which means the photon is at a shorter wavelength than the pumping laser photon. A quantum of thermal energy is thus carried off by a higher energy photon. If this process occurs with high efficiency, the sample can experience net cooling.

Fluorescence and absorption spectra guide the design of a laser cooling experiment. In Fig. 2, the absorption and emission spectra of Tm-doped ZBLAN glass are depicted (ZBLAN is a fluoride glass used to make optical fibers). The vertical line indicates the mean fluorescence wavelength, corresponding to the average photon energy emitted. It is essential that the material be excited with laser photons having smaller energy and hence with longer wavelength than the mean fluorescence wavelength (λ_F). The material absorption, however, has a tendency to decrease significantly at these longer wavelengths. This defines a narrow window of wavelengths where the exciting laser can be tuned to achieve laser cooling. This window is indicated by the shaded anti-Stokes cooling tail in Fig. 2. The wavelength dependence of laser cooling predicted by Fig. 2 is confirmed in experiments. Figure 3 shows laser cooling of Tm:ZBLAN as a function of the pump laser wavelength. Points below the horizontal line represent net cooling; above the line corresponds to heating. The solid curve is predicted behavior based on the spectra. The inset depicts false color thermal images of a sample during such experiments. The dark image indicates sample heating and bright reveals net cooling.[3]

In our experiments, the laser illuminates the sample continuously. The temperature decreases monotonically and eventually attains an equilibrium value. The sample remains cold as long as the laser light is present. Representative dynamics are illustrated in Fig. 4 for a sample of Yb:ZBLAN. The laser beam is turned on at time t = 0 and turned off at t = 3 hours after maximum cooling is obtained. Note the long timescale of the heating recovery process, which indicates that exceptionally good thermal isolation has been obtained between the sample and the chamber in which it is housed. We can quantify the fundamental efficiency of laser cooling based on the preceding discussion. The amount of energy removed by the emitted photons is, on average:

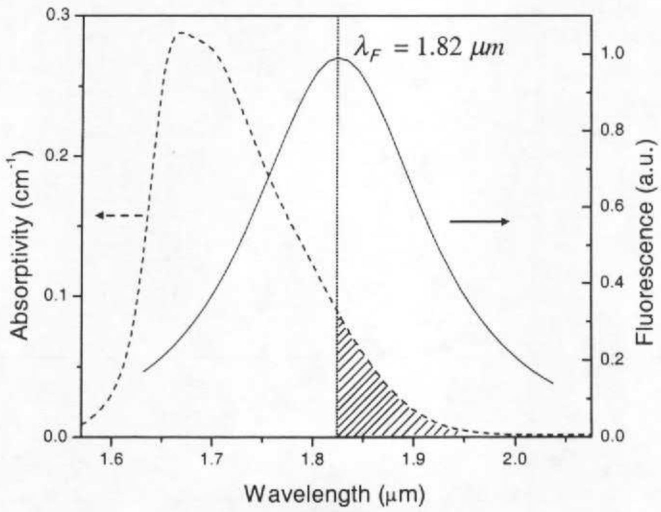

Figure 2. Absorption and fluorescence spectra for Tm-doped ZBLAN glass. The vertical line indicates the mean fluorescence wavelength; shaded region is where cooling occurs.[3]

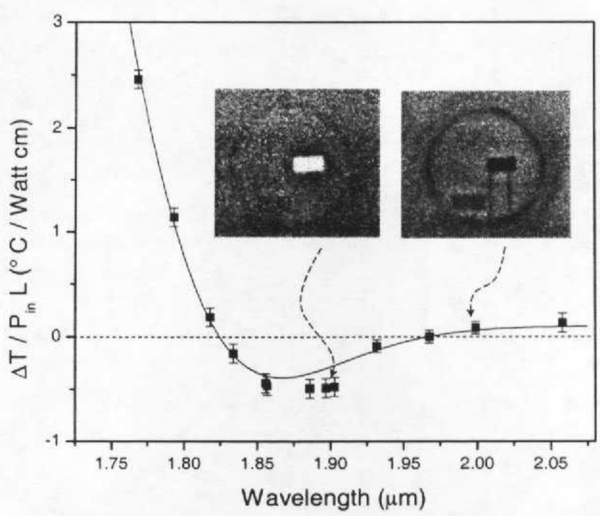

Figure 3. Temperature change of Tm:ZBLAN as a function of pump wavelength. The data are normalized to the laser power. Solid line is model fit based on the spectra in Fig. 2. Inset depicts false color thermal images of cooling (bright) and heating (dark) obtained with an infrared camera.[3]

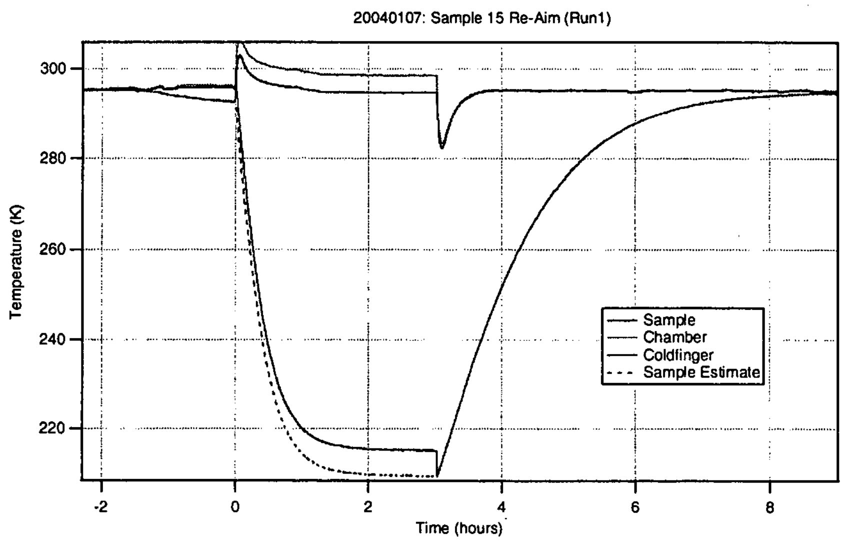

Figure 4. Laser cooling of Yb-doped ZBLAN glass by an amount 88 K. Illumination commences at t = 0 and steady-state is reached after about 3 hours. The upper curves depict thermocouple monitors of the chamber temperature, which is slightly heated by the escaping luminescence. The dotted curve is the deduced sample temperature when corrected for fluorescence heating of the thermocouples.[4]

$h\nu_F - h\nu$, where $h\nu_F = c/\lambda_F$ (c is the speed of light). The cooling efficiency is, in the absence of parasitic heating, the ratio

$$\frac{h\nu_F - h\nu}{h\nu} = \frac{\lambda - \lambda_F}{\lambda} \approx \frac{kT}{h\nu} \tag{1}$$

where we approximate the quantum of thermal energy removed from the sample as $h\nu_F - h\nu \approx kT$. The key point of Eq. (1) is that for a given temperature, greater efficiency can be obtained by designing a cooling system that operates at longer wavelengths. The tradeoff is that the non-radiative lifetime in the upper state tends to get shorter as the emitted photon energy gets smaller. When energy decays in a non-radiative manner, it adds heat to the system and hinders cooling. Selection of materials for laser cooling must carefully consider the efficiency guideline shown in Eq. (1) against deleterious heating associated with non-radiative recombination.

We have demonstrated net laser cooling in Yb-doped ZBLAN glass, crystalline YAG (yttrium aluminum garnett), and Y_2SiO_5. In all these experiments, the laser wavelength is around 1 μm, which is dictated by the absorption spectrum of the Yb atoms in the host material. To test the wavelength scaling of efficiency predicted by Eq. (1), we doped ZBLAN with the rare-earth element thulium (Tm). With Tm-doping, the absorption shifts deeper into the infrared – very close to the wavelength of 2 μm. Results indicate a cooling efficiency about twice as high as Yb-doped systems, confirming the fundamental behavior described by Eq. (1).[3]

The rate of heat removal from the sample is called the cooling power (P_{cool}). The energy partitioning argument used to derive Eq. (1) also leads to a fundamental relationship between absorbed laser power at wavelength λ ($P_{absorbed}$) and radiated cooling power at λ_F:

$$\frac{P_{cool}}{P_{absorbed}} = \frac{\lambda - \lambda_F}{\lambda} \tag{2}$$

which indicates that cooling power improves as the excitation wavelength (λ) increases. This scaling has also been demonstrated experimentally by comparing cooling power obtained with Yb-doped systems at $\lambda \approx 1$ μm to Tm-doped systems at $\lambda = 1.9$ μm.[3]

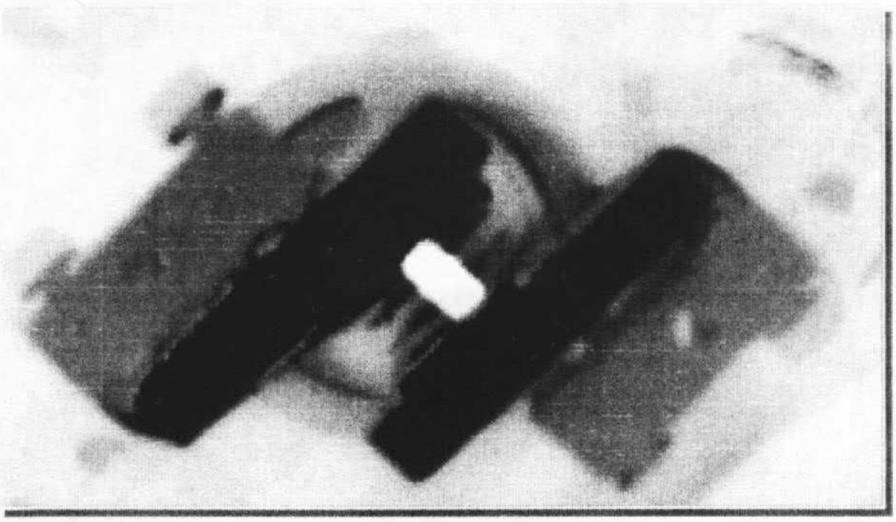

Figure 5. A multi-pass pumping geometry is obtained by placing the glass sample between two mirrors in vacuum. In this thermal image, the cold sample (bright) is clearly seen between the dark silhouettes of the mirror holders. One mirror has a small hole in it to allow entrance of pump laser light.

3. DESIGNING A LASER COOLER

In the discussion of Fig. 2, we indicated that laser pumping must take place at wavelengths longer than the mean fluorescence wavelength. Absorption in this spectral region is weak and a significant amount of pump power can pass through the sample without contributing to cooling. Wasting these pump photons degrades the overall efficiency. To combat this, we have implemented various schemes to recirculate the laser pump power. An approach that has provided dramatic improvements to cooling power is displayed in Fig. 5. By placing the sample between mirrors, a multi-pass geometry is realized. This pump recirculation concept led to record cooling of Tm-doped glass.[5]

To obtain large cooling, the sample must have minimal external thermal loads. Conductive heating is dramatically reduced by supporting the sample in a carriage that is fashioned from small diameter optical fibers or microscope slides. The contact area of the support fixtures is extremely small thereby minimizing conductive heat transfer from the chamber. Support structures with properly chosen glass will not absorb the cooling fluorescence. Convective heat currents are eliminated by evacuating the sample chamber. Thermal radiation from the sample walls can overwhelm the cooling radiation in the spectral region around λ_F. This problem is mitigated by coating the chamber walls with a material that has low emissivity at thermal wavelengths near 10 microns.[4] We have recently obtained net cooling using the host BaY_2F_8 doped with Tm. This host is expected to be mostly transparent to thermal wavelengths and may ease the design of the sample chamber.

A conceptual schematic of a practical laser cryocooler is depicted in Fig. 6. It is based on rare-earth-doped glass, pumped with a compact laser diode. Laser light is routed to the sample with an optical fiber. The ends of the sample are mirrored to achieve multiple round trips of the laser light to maximize the absorbed power. Laser light couples to the cooling element through a small hole in one of the mirrored surfaces as shown. The load (i.e. cold finger) is in thermal contact with one of the mirrored surfaces. Cooling fluorescence power is absorbed on the chamber walls. One of the primary challenges in implementing such a system is attaining high efficiency, low absorption mirror coatings. Any heating that occurs in the mirrors can harm and even overwhelm the desired fluorescence cooling.

Another critical design issue is photon waste management. The cold object such as an infrared sensor must be thermally (i.e. conductively) connected to the cooling element, yet shielded from the emitted fluorescence. Metallic mirrors are absorptive and would generate too much heat. High reflectivity dielectric coatings do not

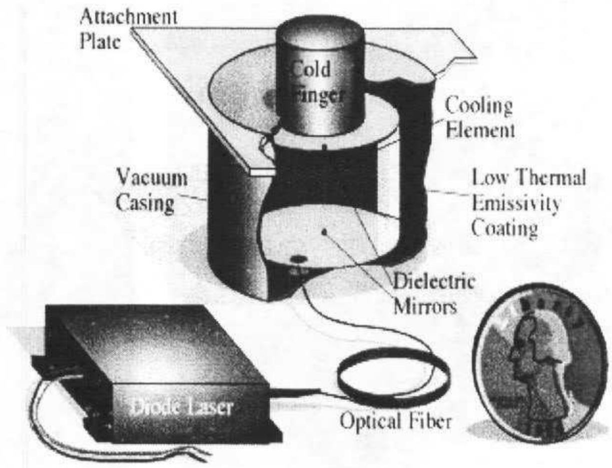

Figure 6. Schematic of a compact laser-cooler that could be used in an infrared sensor application.

give adequate reflectivity at large incidence angles and therefore do not block the isotropic fluorescence. We are designing photonic bandgap interfaces to solve this problem.

4. SEMICONDUCTORS: THE NEXT GENERATION OF LASER COOLERS

Laser cooled semiconductors offer the potential for new cryocooler applications involving sensors and electronics. The physical principles in play are essentially the same as already described, but semiconductors have an important advantage compared to doped glasses: they can cool to below 10 K.[6] In semiconductors, the electron population in the lower energy level is always maintained, even at absolute zero. It is much more challenging to attain this condition in glass. This feature of semiconductors could allow absolute temperatures below 10 K to be achieved. Very small amounts of semiconducting material can provide an enormous amount of pump laser absorption. This means that much more compact and efficient devices can be designed. The technology for semiconductor device fabrication is advanced, mature, and potentially inexpensive in production.

Figure 7 depicts results of a luminescence experiment with a GaAs semiconductor structure. When pumped with laser light at wavelength 890 nm, the luminescence peak is shifted to a wavelength of about 875 nm. This is one of the desired conditions for laser cooling – radiation occurring at predominantly shorter wavelengths than the pump has the potential to remove heat from the system.

The device used to generate the data in Fig. 7 has been passivated with the alloy InGaP.[7] Both top and bottom surfaces of the bulk GaAs sample have thin layers of InGaP grown on them. Passivation is necessary to stop the near instantaneous recombination of excited electrons that occur at the semiconductor-vacuum interface. If the excitations disappear before they can be thermally excited by the semiconductor, radiative cooling is completely suppressed. The energy barrier at the InGaP-GaAs interface prevents charge carriers in GaAs from reaching the surface. The thin InGaP layers present a negligible amount of absorption to the pump laser and escaping luminescence. The process of metal-organic chemical vapor deposition (MOCVD) of InGaP has been shown to produce the lowest interface recombination velocity for bulk GaAs. In addition to having proper surface passivation, the bulk semiconductor must be extremely pure and defect free to prevent the decay of laser-excited electrons by a non-optical pathway, which would lead to heating.

There are still serious challenges that must be overcome before net cooling of semiconductors is realized. The most important obstacle is luminescence radiation trapping. Unlike transparent glasses and crystals, semiconductors generally have an index of refraction that is much greater than 1. Since there is a substantial index

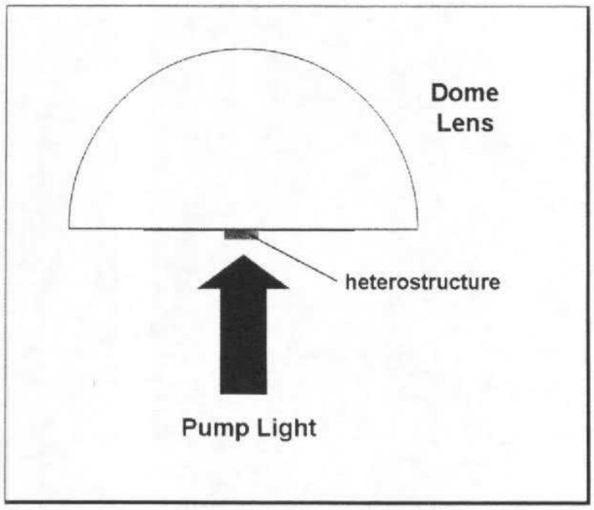

Figure 8. Luminescence removal by use of a nearly-index matched dome lens. The passivated heterostructure is placed in optical contact with the dome as shown and pumped from the opposite side. The lens must have extremely low absorption to prevent conductive re-heating by the escaping luminescence.

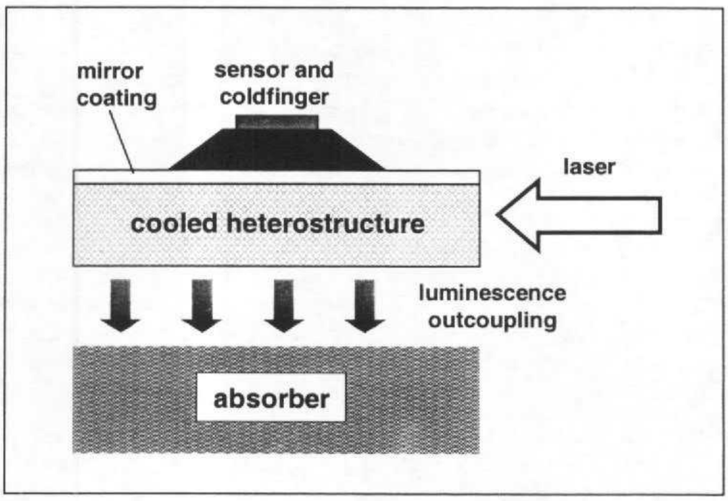

Figure 9. Nanogap concept for luminescence removal. When the heterostructure is brought to within a fraction of a wavelength of an absorbing substrate, total internal reflection can be overcome. Radiation power is transferred to the absorber via evanescent waves that leak across the gap. The gap is held at vacuum to minimize convective heat transfer between the two layers.

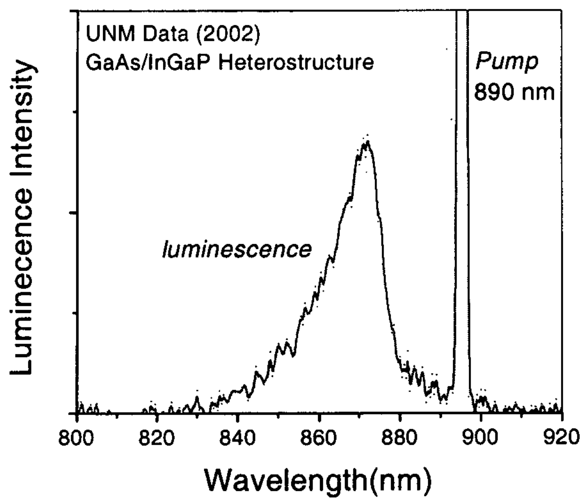

Figure 7. Blue-shifted luminescence of bulk GaAs when pumped at the band edge (890 nm). To attain net cooling, the luminescence must be removed from the semiconductor heterostructure with high efficiency, which presents a challenging engineering problem.

mismatch between the semiconductor and vacuum, most of the luminescence tends to be trapped by total internal reflection. This trapping is extremely deleterious to cooling. Efficient extraction of luminescence is therefore essential to obtain net cooling.

One method to address radiation trapping is to place a high index, low absorption lens on one surface.[8] This concept is depicted in Fig. 8. Dome lenses are widely used in the design of light emitting diodes. In a laser cooling application, the lens must present minimal absorption to the escaping light; otherwise it would heat up and transfer thermal energy back to the semiconductor by conduction. In general, the closer the refractive index of the lens matches the semiconductor, the higher its absorption. Careful measurement of absorption in candidate lens materials has allowed us to identify ZnS and ZnSe as the best choices for a laser cooling application.[9] A dome lens fabricated from either of these materials would allow about 22% of the luminescence to escape from the passivated GaAs structure. This is anticipated to be sufficient outcoupling to realize net cooling.

Potentially greater luminescence removal can be effected by implementing the "nanogap" concept. This relies on the principle of frustrated total internal reflection. The idea, depicted in Fig. 9, operates as follows[10]: An absorbing substrate is placed an appropriate distance from the semiconductor-vacuum interface; this interface normally exhibits total internal reflection. When this distance is a fraction of a wavelength of light, however, coupling of optical power between the two substrates can take place. Total internal reflection is partially overcome and luminescence can couple to the absorbing substrate (e.g. silicon). In this way, the heterostructure and absorber are optically coupled yet thermally isolated.

We have calculated that a nanogap can provide more than twice the luminescence removal of a dome lens. The challenge, of course, is placing the two materials close enough that they optically couple without significant thermal conductivity between them. The necessary distance for optical coupling is less than 100 nm and calls for extreme techniques of nano-fabrication. We register the gap using a grid of nanoposts, an example of which is depicted in the scanning electron microscope image of Fig. 10. This shows an oxide post on a silicon absorber; a flat GaAs heterostructure would be bonded to an array of such posts. Design of the post structure is critical in both size and number. The height must be small enough to maintain the required gap and the cross section (diameter) of the posts must minimize thermal conductivity between the absorber and cooling layer. The spatial separation of the posts must be sufficient to give structural support to the thin, cooling layer. We are currently engaged in research to implement the nanogap technique in laser cooling of semiconductors.

Figure 10. Scanning electron microscope image of a post for registering the nanogap separation. This post is made of silicon dioxide fabricated on a silicon substrate. The height of the post is less than 50 nm (note 100 nm reference bar).

Two of us (Sheik-Bahae and Epstein) have made a theoretical investigation of laser cooling in semiconductors and GaAs in particular.[6] It was determined that there are no fundamental physical or engineering issues to prevent realization of net laser cooling. Of particular interest is the significantly enhanced performance that is predicted to occur as the sample temperature decreases below 300 K. Just as in laser cooling of doped glass, minimizing the rate of non-radiating, heat producing energy decay is essential. The role of radiation trapping was also quantified in this model.

5. SUMMARY

We have presented a brief overview of our research on laser cooling of solids. The physics of laser cooling in rare-earth doped glasses and crystals is now well understood. This knowledge base drives the design and engineering of advanced laser cryocoolers for infrared sensors. Record amounts of temperature decrease, cooling power, and efficiency have been achieved very recently. Our analysis shows that direct laser cooling of semiconductor crystals such as GaAs is possible. This could allow the eventual implementation of integrated, miniature cryocoolers in compact sensors and electronics.

REFERENCES

1. R.I. Epstein, M.I. Buchwald, B.C. Edwards, T.R. Gosnell, and C. Mungan, "Observation of laser-induced fluorescent cooling of a solid", *Nature* **377**, 500-502, 1995.
2. P. Pringsheim, *Z. Phys.* **57**, 739-746, 1929.
3. C.W. Hoyt, M. Sheik-Bahae, R.I. Epstein, B.C. Edwards, and J.E. Anderson, "Observation of Anti-Stokes Fluorescence Cooling in Thulium-Doped Glass", *Phys. Rev. Lett.* **85**, 3600-3603, 2000.
4. J. Thiede, J. Distel, S. Greenfield, R.I. Epstein, manuscript in preparation.
5. C.W. Hoyt, M.P. Hasselbeck, M. Sheik-Bahae, R.I. Epstein, S. Greenfield, J. Thiede, J. Distel, and J. Valencia, "Advances in laser cooling of thulium-doped glass", *J. Opt. Soc. Am. B* **20**, 1066-1074, 2003.
6. M. Sheik-Bahae and R.I. Epstein, "Can Laser Light Cool Semiconductors?", *Phys. Rev. Lett.* **92**, 247403-1–247403-4, 2004.
7. J.M. Olson, R.K. Ahrenkiel, D.J. Dunlavy, B. Keyes, and A.E. Kibbler, "Ultralow recombination velocity at $Ga_{0.5}In_{0.5}P$/GaAs heterointerfaces", *Appl. Phys. Lett.* **55**, 1208-1210, 1989.

8. H. Gauck, T.H. Gfroerer, M.J. Renn, E.A. Cornell, and K.A. Bertness, "External radiative quantum efficiency of 96% from a GaAs/GaInP heterostructure", *Appl. Phys. A* **64**, 143-147, 1997.
9. B. Imangholi, M.P. Hasselbeck, and M. Sheik-Bahae, "Absorption spectra of wide-gap semiconductors in their transparency region" *Opt. Commun.* **227**, 3337-341, 2003.
10. R. I. Epstein, B. C., Edwards, and M. Sheik-Bahae, "Semiconductor-based optical refrigeration", U. S. Patent 6,378,321 (2002).

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