LA-UR-95- 3268

CONF-9509236--

TITLE: DEVELOPMENT OF A FLUORESCENT CRYOCOOLER

AUTHOR(S): Bradley C. Edwards, NIS-2
Melvin I. Buchwald
Richard I. Epstein, NIS-2
Timothy R. Gosnell, MST-10
Carl E. Mungan, MST-10

SUBMITTED TO: AIAA Satallite Conference, September 20, 1995, Utah

MASTER

Los Alamos
NATIONAL LABORATORY

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Form No. 836 R5
ST 2629 10/91

DISCLAIMER

Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.

DEVELOPMENT OF A FLUORESCENT CRYOCOOLER

Bradley C. Edwards, Melvin I. Buchwald, Richard I. Epstein,
Timothy R. Gosnell, and Carl E. Mungan

Los Alamos National Laboratory
Los Alamos, New Mexico

Abstract

Recent work at Los Alamos National Laboratory has demonstrated the physical principles for a new type of solid-state cryocooler based on anti-Stokes fluorescence. Design studies indicate that a vibration-free, low-mass "fluorescent cryocooler" could operate for years with efficiencies and cooling powers comparable to current commercial systems. This paper presents concepts for a fluorescent cryocooler, design considerations and expected performance.

Introduction

Recent laboratory measurements have demonstrated laser-induced, anti-Stokes fluorescent cooling of a solid[1]. Combining this laboratory work with computer simulations and information on available technology, we estimate that a practical, first-generation, all-solid-state fluorescent cryocooler will have the following properties:

- no vibrations
- does not produce and is not susceptible to electromagnetic interference
- cool to 77 K
- be ~1% efficient (DC power to cooling power)
- weigh less than 3 kg/Watt
- have a lifetime of 10 years continuous operation.

This first-generation cryocooler will employ material with a demonstrated fluorescent cooling capability (ytterbium doped ZBLANP). With improved cooling materials and designs future cryocoolers could be over twice as efficient at 77 K and cool to lower temperatures.

Anti-Stokes Fluorescent Cooling

Anti-Stokes fluorescence is the phenomena in which a substance that is excited by radiation at one wavelength fluoresces at a *shorter* wavelength. This results in more energy being radiated than being absorbed for each photon. Utilizing this processes for cooling may, at first, appear to violate common sense: a multi-watt laser is focused into a mostly transparent material, and the material cools. A closer look
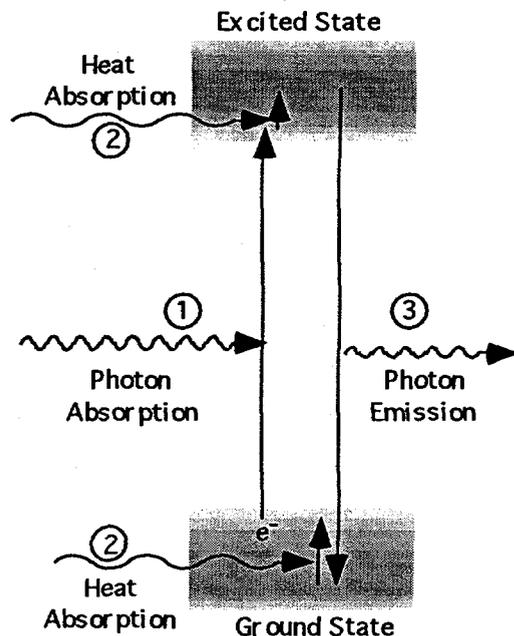
Figure 1: A diagram of the atomic processes that produce fluorescent cooling. 1) Electrons in the upper levels of the ground-state manifold are pumped to the lower levels of the excited-state manifold. 2) The populations of the energy levels re-equilibrate through phonon absorption. 3) Fluorescent photons (more energetic than the pump photons) are emitted as the ions de-excite back to the ground state. Each phonon absorption extracts heat from the system.

1

at a specific cooling process shows how fluorescent cooling works. Consider a glass doped with a rare-earth ion that has broad ground and excited states (Fig. 1). Each broad energy state or manifold consists of several closely spaced energy levels that are in thermal equilibrium among themselves (since we will be discussing glasses the energy levels are actually inhomogeneously broadened bands but for simplicity we will refer to them as discrete levels). When ions are radiatively pumped from the top of the ground-state manifold to the bottom of the excited-state manifold (process 1), the thermal equilibrium in each manifold is upset. The upper level of the ground-state manifold is underpopulated and the lower level of the excited-state manifold is overpopulated. Both manifolds quickly return to equilibrium by absorbing phonons (heat) from the surroundings (process 2). At some later time the ions de-excite by emitting photons (process 3). The fluorescent photons remove both the pump photon energy and the absorbed phonon energy from the

system. The net result of this process is the removal of heat from the material.

In the ideal case, where there are no nonradiative relaxations from the excited- to the ground-state manifolds, there will be one fluorescent photon for each pump photon and each fluorescent photon carries off, on the average, thermal energy equal to the difference between the pump photon energy and the mean fluorescent-photon energy. The fluorescent cooling power, P_{cool}, is thus proportional to the absorbed pump power, P_{abs}, and to the average difference in the photon energies of the pump and fluorescence radiation. In terms of wavelength, the cooling power can be written as

$$P_{cool}(\lambda) = P_{abs}(\lambda)(\lambda - \lambda_F)/\lambda_F, \qquad (1)$$

where λ is the wavelength of the incident pump radiation and λ_F is the wavelength corresponding to the mean of the fluorescent photons energy.
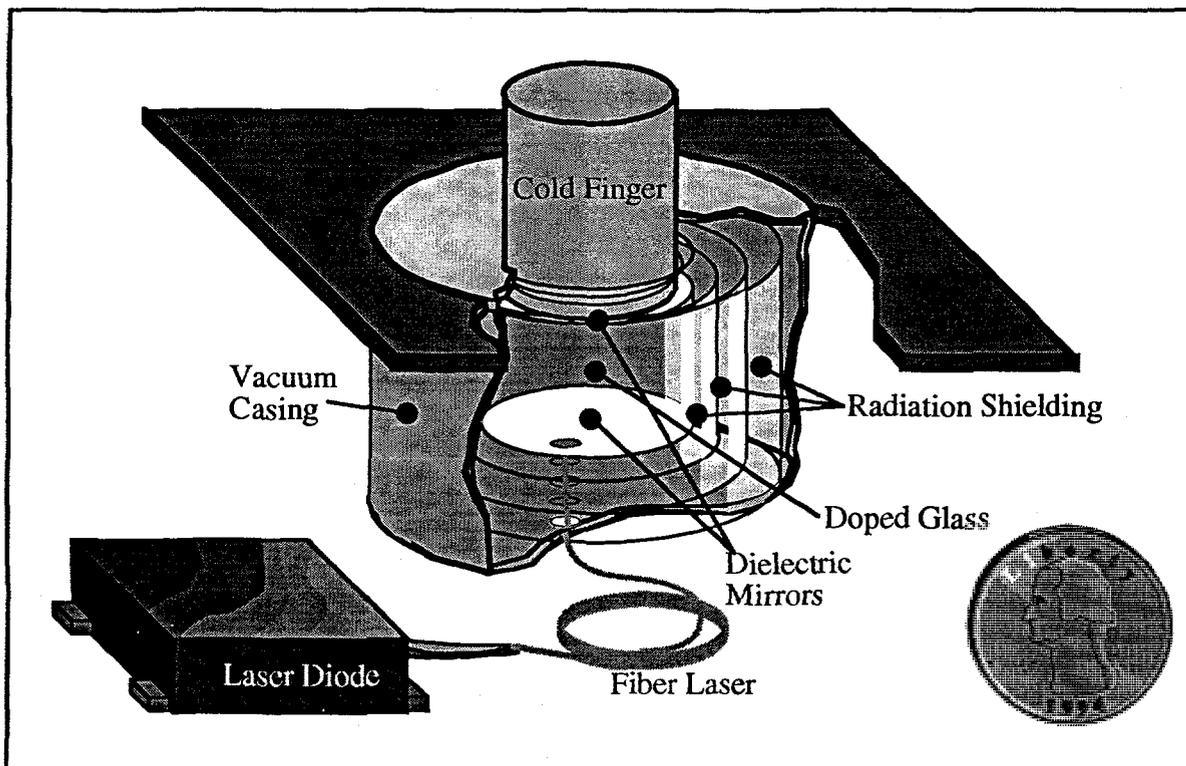

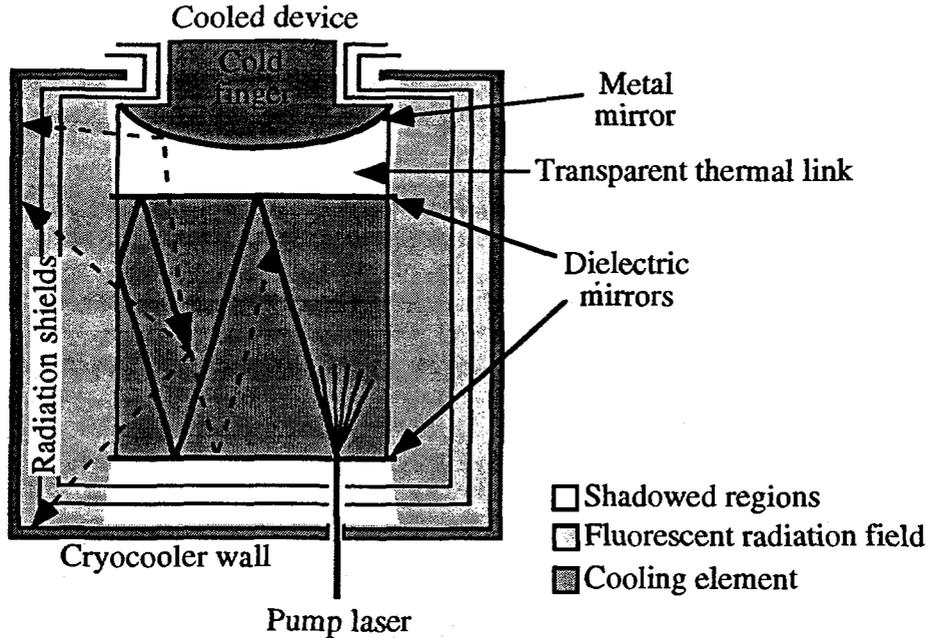Figure 2: A design for a fluorescent cryocooler.

2

Figure 3: Schematic of the cooling element and cold finger design. The input laser light is shown as solid lines and the fluorescence as dashed. Light gray areas show where the fluorescent radiation field is most intense. White regions are regions in the shadow of the dielectric mirrors. These shadowed regions are not completely free of fluorescent radiation due to the bandpass of the dielectric mirrors so an additional metal mirror and baffles are required to eliminate any fluorescent radiation loading on the object to be cooled.

First-Generation Fluorescent Cryocooler

Overall Design

An especially interesting application of a solid-state fluorescent cryocooler is for use in space where performance attributes are critical. In these applications fluorescent cryocoolers would be a valuable asset because they are vibration-free, low-mass, rugged, long-lived and efficient.

Figure 2 illustrates one design for a fluorescent cryocooler that might be used for space applications. This design is based on using ytterbium (Yb^{3+}) doped heavy metal fluoride glasses for the fluorescent cooling element. Preliminary studies have demonstrated that one such material, $ZBLANP:Yb^{3+}$, is an effective fluorescent cooler[1].

Our initial design utilizes a combination of diode lasers coupled with an Yb-fiber laser to efficiently produce pump radiation at the desired wavelength (see Fig. 2). The fiber carrying the pump radiation passes through the wall of the cryocooler and to a cylindrical block of $ZBLANP:Yb^{3+}$, the cooling element at the heart of the device (see Fig. 3). Dielectric mirrors that reflect the pump radiation are deposited on both ends of the cooling element. The pump radiation enters the cooling element through a small hole in one of the mirrors. The pump radiation is largely beamed along the axis of the cylinder and is repeatedly reflected from the dielectric mirrors and from the sides of the glass cylinder (by total internal reflection). Ultimately, the pump radiation is absorbed by Yb^{3+} ions, and the doped glass cools as it fluoresces. Most of the isotropic fluorescent radiation escapes from the cooling element and is absorbed by the warm walls of the cooling chamber. On the back of one of the dielectric mirrors will be attached a transparent connection and metal mirror to create a completely shadowed region where the cold finger can be attached.

Each of the components of the fluorescent cryocooler must be precisely combined to

produce an effective device. We now examine some the considerations involved in designing the key components and how these choices can affect the overall performance of the cooler.

Cooling Element Composition:

The two essential requirements for a fluorescent cooling element is that it exhibit anti-Stokes fluorescence and that the resulting radiation-induced cooling is not severely degraded by heating from nonradiative decays. Both of these requirements are well satisfied by ZBLANP:Yb^{3+}.

The Yb^{3+} ion has a simple energy level structure consisting of only a ground-state and a single excited-state manifold at ~1000 nm. The room temperature fluorescence and absorption spectra of ZBLANP:Yb^{3+} shown in Fig. 4 indicate that anti-Stokes fluorescence is achievable with this material. Since the mean energy of the fluorescent photons is at λ_F =

995 nm, whenever the excitation is pumped with radiation where $\lambda > 995$ nm, the glass cools (see Eq. 1). The absorption spectrum shows that it is indeed possible to pump ZBLANP:Yb^{3+} at these long wavelengths.

It only takes a few phonon-emitting, nonradiative decays between the excited-state and ground-state manifolds to overwhelm the fluorescent cooling effect. In pure ZBLANP:Yb^{3+} this is not a problem. The radiative lifetime of the upper manifold is ~2 ms, whereas the nonradiative decay lifetime, which involves the simultaneous emission of more than 15 phonons, is estimated to be much greater than a year. If multi-phonon emission were the only nonradiative process essentially all of the decays would occur radiatively.

Nonradiative transitions can be a problem, however, when contaminants are present. An excited Yb^{3+} ion can de-excite by transferring its energy to a nearby contaminant ion such as
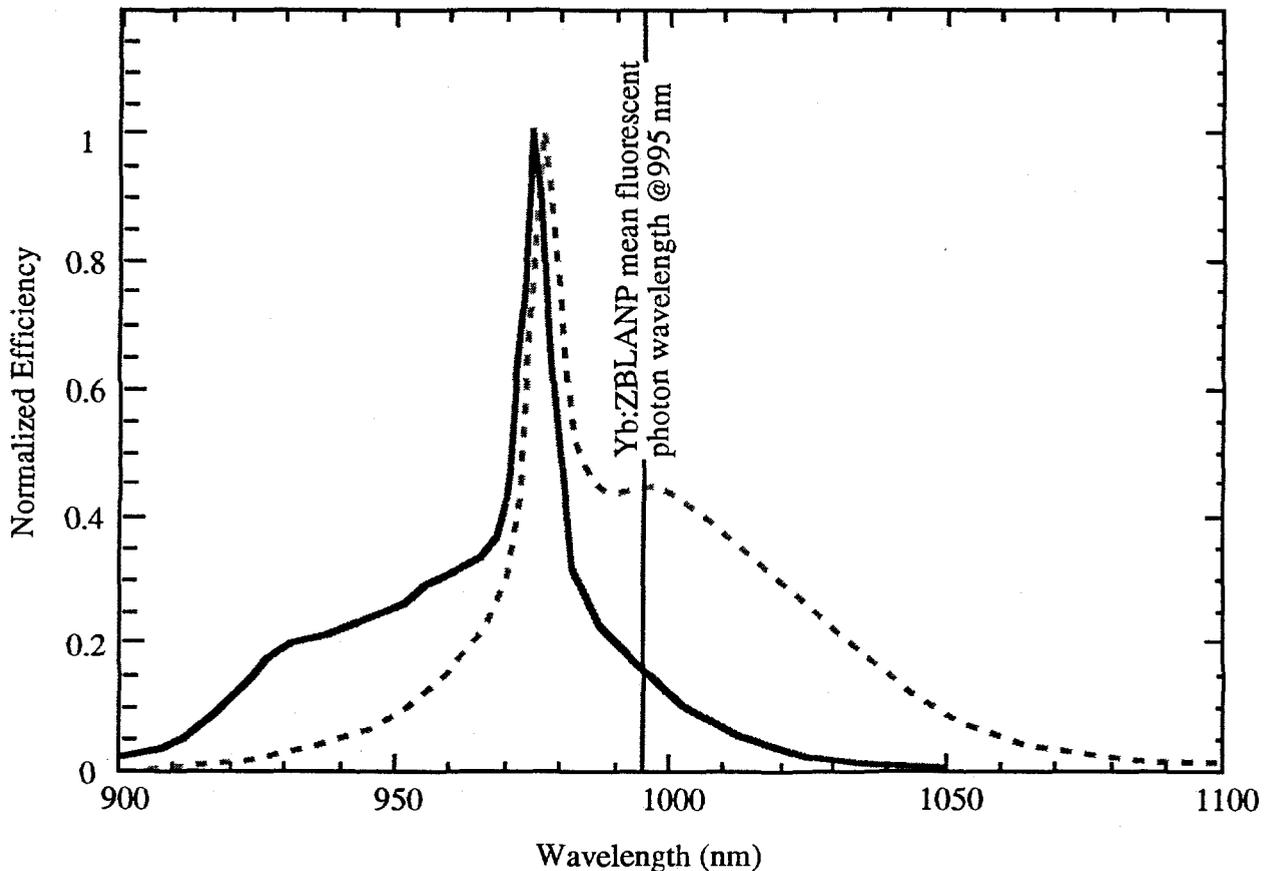

Figure 4: Fluorescence and absorption spectra for Yb^{3+} in ZBLANP.

Er^{3+}, Dy^{3+}, Sm^{3+}, Fe^{3+}, etc. These impurity ions would quickly decay nonradiatively producing heat. The severity of this problem is amplified by excitation migration in which excitations move among neighboring Yb^{3+} until it is quenched by an impurity. Extremely pure materials are needed to produce an effective cooling element.

The absorption and fluorescence spectra vary with temperature since they depend on the excitation populations of each level in both manifolds. At low temperatures the populations of upper energy levels of the ground-state manifold decrease thereby reducing the absorptivity at long wavelengths. Similarly, the decrease of the population of the upper levels of the excited-state manifold at low temperatures reduces the fluorescence at

short wavelengths. As one might expect, the net effect of a decreased temperature is that it is more difficult to achieve anti-Stokes fluorescent cooling.

We have simulated the temperature response of ZBLANP:Yb^{3+} using measured energy levels, and measured absorption and fluorescence spectra. Assuming a Boltzmann distribution for the population of each level in each manifold we modeled the widths and transition probabilities for transitions between each of the states. Our calculations reproduced the observed room-temperature fluorescence and absorption spectra and simulated the behavior at other temperatures. We found that as the temperature decreases, the mean fluorescence shifts to longer wavelengths and the absorption spectrum shifts to shorter
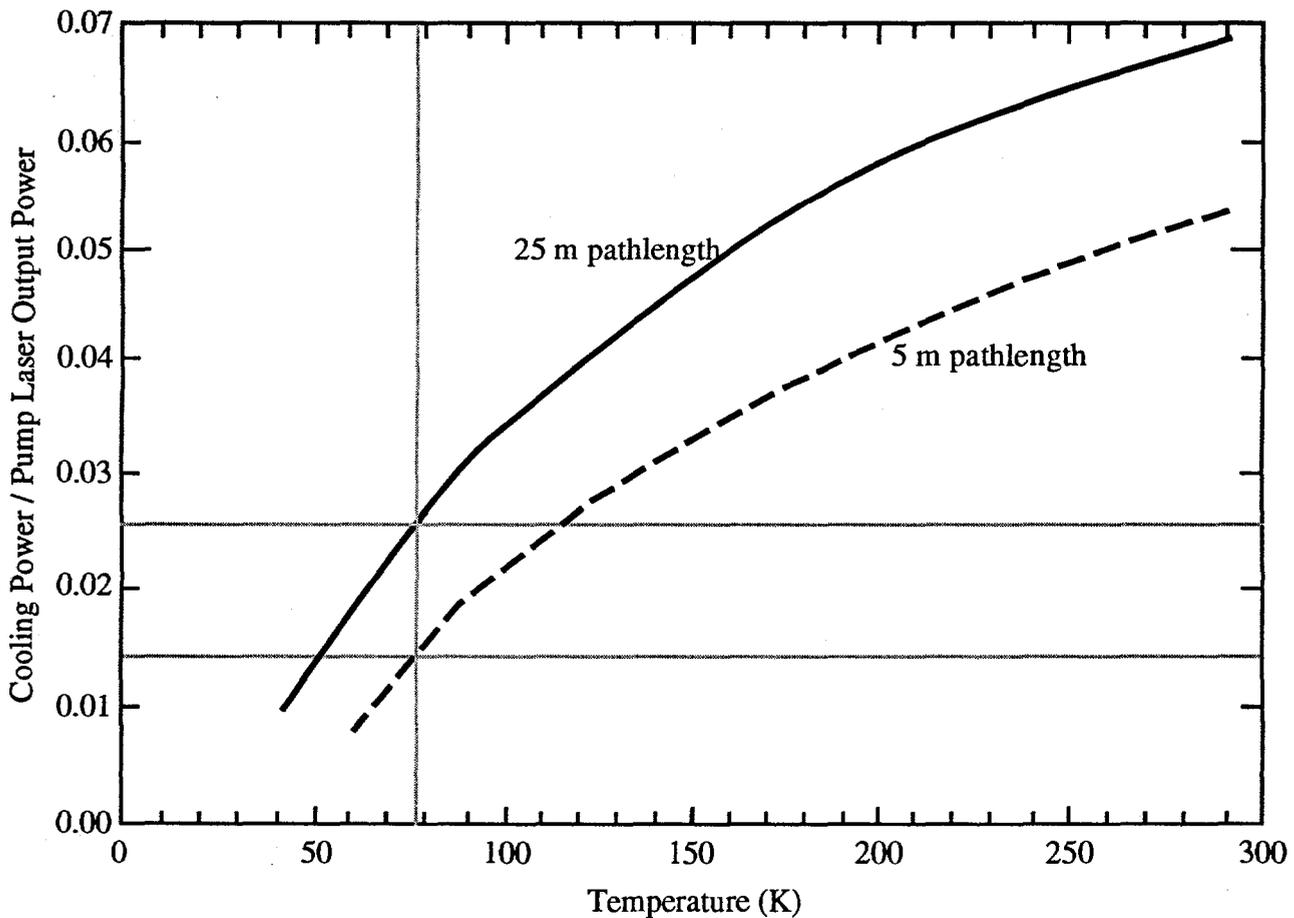

Figure 5: Calculated cooling efficiency vs. temperature for a 5 m and a 25 m pathlength of 2% Yb^{3+} doped ZBLANP .

wavelengths. While these two effects reduce the anti-Stokes fluorescence, useful cooling efficiency can be achieved with ZBLANP:Yb^{3+} at temperatures as low as 50 K (see Fig. 5).

Radiation Trapping:

The primary considerations in designing the cooling element, besides the material choice are geometrical. The cooling element must completely absorb the pump radiation while allowing the fluorescent radiation to escape.

To efficiently utilize the pump laser power, the radiation should have the longest possible wavelength (see equation 1) and it should be completely absorbed in the cooling material. Since the absorptivity decreases at long wavelength, satisfying these two conditions requires that the radiation be trapped in the cooling element until it is absorbed. In the first-generation fluorescent cryocooler design we use dielectric mirrors on the ends of the cooling element to trap the pump radiation. As Fig. 3 shows, the laser radiation enters the cooling element in a narrow cone aligned with the axis of the cylinder. The beaming is sufficiently narrow so that the light is almost perfectly reflected from the dielectric mirrors at the ends or by total internal reflection from the sides of the cooling element. Initial tests indicate that this trapping scheme is effective. The trapping efficiency is determined by the quality of the glass, mirror construction and the ratio of the mirror area to the area of the entrance hole through which the pump light enters. Several practical considerations determine the currently attainable trapping efficiency:

- Dielectric mirrors can be made that reflect 99.97% of the incident radiation for angles within 30 degrees of normal. This is adequate for exit angles of standard fibers (N.A. = 0.22, exit angles $<\pm 13°$).

- Because of the entrance hole, the light leakage per reflection is equal to the ratio of the area of the hole to the total mirror area. The current design uses a 0.1 mm diameter hole and a 30 diameter mirror to give us a leakage per reflection ratio of 1.1e-5.

- Imperfections in the cooling element or the mirrors may scatter some radiation out of the "trapping cone" before it is absorbed.

We estimate that with current technology the pump radiation can be trapped for more than several hundred passes (~5 m pathlength in our design) and possibly over 1000 passes (~25 m pathlength) through the sample. As can be seen in Fig. 5 this is a critical design consideration and continuing studies on trapping mechanisms are needed.

The fluorescent radiation is emitted isotropically in the cooling element, and most of it is able to escape without any reflections. About 25% of the fluorescence will be trapped by internal reflections and by the dielectric mirrors and will be reabsorbed in the Yb^{3+}. During subsequent fluorescence most of this radiation will escape. This repeated absorption will not seriously degrade the performance of the system because the absorbed and re-fluoresced light have similar mean energies.

Mass of Cooling Element and Cooler:

The maximum heat the fluorescent cooler could extract depends on the mass of the cooling element, the Yb^{3+} concentration and the operating temperature. As an example, Fig. 6 gives the required mass of the cooling element for a 0.5 Watt unit with 2 wt% concentration of Yb^{3+}. For the fluorescent cryocooler to operate at 77 K, the cooler element would have to be at least 80 g; this would corresponding to a cylindrical element with a height and diameter of 3 cm.

The remaining mass of the cryocooler is composed of the outer wall, diode lasers, heat sinks and control electronics. The outer wall must be sufficient to transport the waste heat away. Similarly, heat sinks must be implemented to remove the heat generated by the diode lasers. These thermal conduction paths would be constructed from diamond coated aluminum or copper. The outer wall
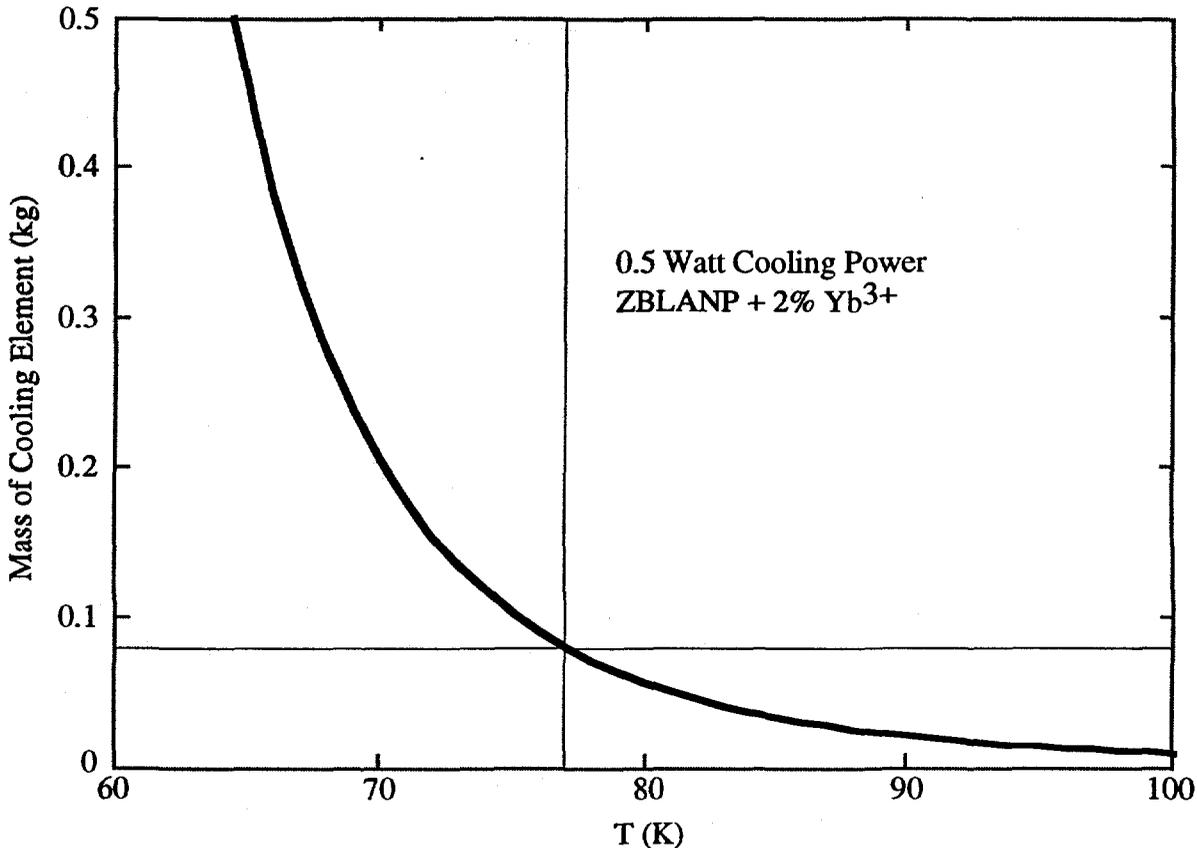
Figure 6: The minimum mass of the cooling element of a 0.5-watt, ZBLANP:Yb^{3+}-based fluorescent cryocooler

mass would be roughly 350 gms depending on the design. Each laser diode set (8 for a ten year lifetime) and heat sink would have a mass of up to 100 gms. The electronics to operate the laser diode could be very simple in a spacecraft system where DC power is readily available and may account for one board or roughly 300 gms. The total mass estimate for a 0.5 watt fluorescent cryocooler is thus about 1.5 kgs for a first-generation system. By integrating the cryocooler into the overall spacecraft design, a cryocooler system might be produced with a mass as small as several hundred grams.

Pump Radiation Source:

A cryocooler for use in space applications requires a radiation source that is compact, efficient, reliable and rugged and has the needed power and wavelength. Fortunately, recent developments in diode lasers and solid state lasers have made a number of options

available. The radiation source we are currently considering is a diode pumped Yb-fiber laser.

To produce 0.5 W of cooling power at 77 K, the cooler needs ~20 W of laser light at 1020 nm. Commercially available high efficiency diode lasers can produce tens of Watts of cw power at 980 nm with ~60% efficiency (DC power to light). Yb-fiber lasers have been made that can convert the 980 nm radiation to 1020 nm radiation with ~80% efficiency[2]. Allowing for an ~80% efficiency in the optical couplings, we need ~50 W of electrical power to run the 0.5 W cooler. This means that for the first-generation fluorescent cryocooler the DC power to cooling power efficiency would be ~1%.

The lifetime of the fluorescent cryocooler will be limited by the lifetimes of the diode lasers. Currently commercial dioe lasers have mean times to failure of ~14 months. Cooler

7

lifetimes can be extended by combining multiple, redundant, relatively low-power diode lasers. This would have the added advantage of distributing the heat load. For a system to achieve a ten year lifetime 8 sets of diodes would be required.

Shielding and Shadowing:

An important part of the design of an effective fluorescent cryocooler is controlling the flow of fluorescent radiation and the thermal radiation from the warm walls of the cooler.

To decrease the heat load on the cooling element from the thermal radiation of the walls, layers of shielding are interspersed between these components (see Figs. 2 and 3). The shielding material must be transparent to the fluorescence allowing the radiation to escape from the cooling element and be absorbed on the walls. On the other hand, the shields should reflect or absorb the thermal radiation to impede it from reaching the cooling element. One possibility for shielding material would be silica glass coated with a thin layer of titanium dioxide. Since titanium dioxide is highly reflective from 8 to >13 microns[4], these shields would be reflective on the outer surface and roughly blackbodies on the inner surface (silica) to thermal radiation. Shields of this type would reduce the radiative loading by roughly (1 − reflectivity). For example, a 60% reflecting surface on three shields would reduce the heat load by a factor of ~15.

The cold finger and especially the cooled instruments have to be well protected from both the fluorescent and thermal radiation fields. Placing a curved metal mirror behind and in thermal contact with one of the dielectric mirrors would create an area completely shadowed from the fluorescence. (see Fig. 3). The object to be cooled would be attached to this dark area.

Future Directions

Future generations of fluorescent cryocoolers should benefit from improved cooling element materials and from new developments in diode lasers.

Cooling Element Composition:

The design of the first-generation fluorescent cryocooler employs an Yb^{3+}-doped glass which has an exceeding simple energy level structure. However, numerous other dopant-host combinations are possible. Figure 7 illustrates the energy levels of some of the interesting rare-earth ions that are potential fluorescent coolants.
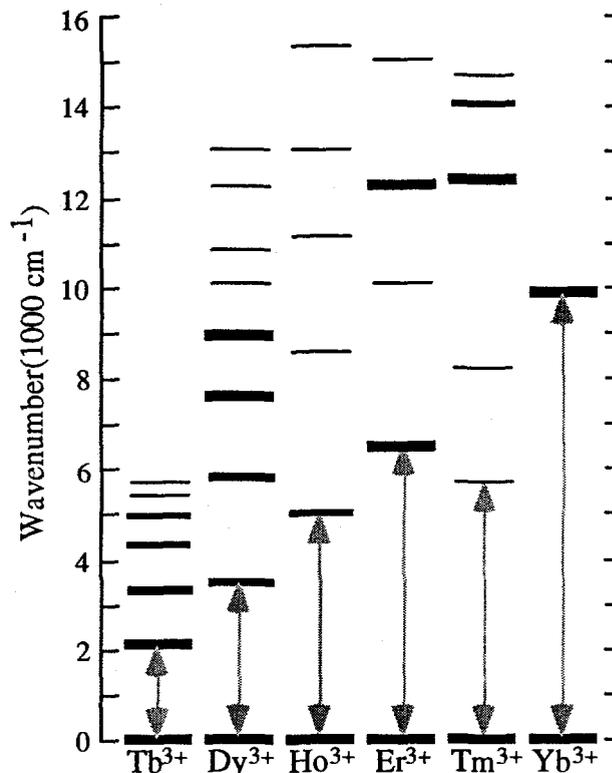

Figure 7: Energy level diagram for several rare-earth ions[3]. The gray arrows show the transitions that might be used for cooling.

The potential cooling transitions must have the property that the radiative decay rate (lifetimes can be on the order of milliseconds for rare-earth ions) must be much faster than the multiphonon emission rate (which depends on the host material, see Fig. 8). Dopant-host candidates such as $ZBLANP:Tm^{3+}$ and $LaBr_3:Dy^{3+}$ may be promising cooling combinations due to their radiative and phonon properties (see Figs. 7 and 8). Due to the lower energy photons required to pump these ions and the width of the states, these combinations should have higher overall

8

cooling efficiency than the ZBLANP:Yb^{3+}. Since the width of the Tm^{3+} states are roughly the same as those for Yb^{3+} (≈ 500 cm^{-1}) the overall efficiency would increase by the ratio of the energy splitting between the ground- and excited-state manifolds.

Recent developments in laser diode technology may make higher efficiency sources at more wavelengths readily available[5]. In particular InGaAsP-based high-power laser diodes have the potential of > 10 year lifetimes. Additionally these diodes may have enough wavelength flexibility to pump an Yb^{3+}-based fluorescent cryocooler without requiring an intermediate Yb-fiber laser.
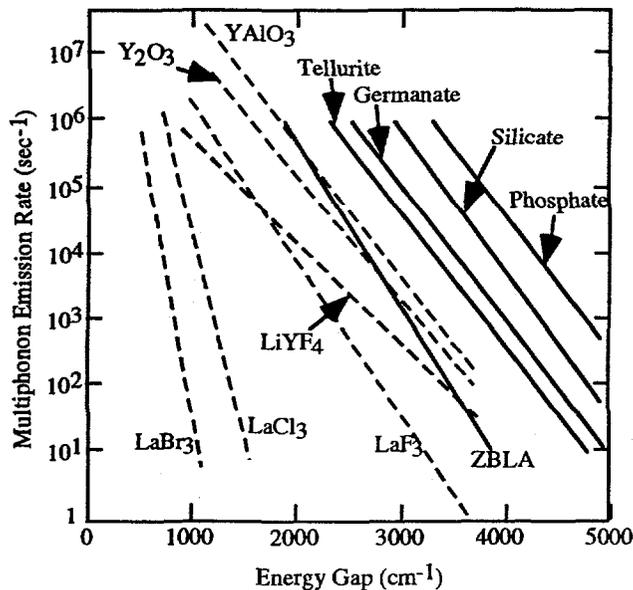

Figure 8: Phonon emission rate vs. energy gap for several glasses (solid lines) and crystals (dashed lines) [6] at room temperature.

Conclusions

Laboratory data has proven the feasibility of cooling macroscopic, solid objects by anti-Stokes fluorescence. We have studied the possibility of using this phenomenon to produce a practical cryocooler. With current technology a fluorescent cryocooler that generates ~0.5 watts of cooling at 77 K seems feasible. This first-generation cryocooler would be ~1% efficient, induce no vibrations, have a long life, and a mass of 1.5 kg. Future systems could be at least twice as efficient and

be usable at much lower temperatures. A small cryocooler of this sort would be well suited for operation in space.

References

1. Epstein, R. I., Buchwald, M. I., Edwards, B. C., Gosnell, T. R., and Mungan, C. E., submitted to *Nature*, 1995.
2. Hanna, D. and Tropper, A. *Laser Focus World* ,123, (May 1995)
3. Macfarlane, R. M. and Shelby, R. M., *Spectroscopy of Solids Containing Rare Earth Ions*, eds A. A. Kaplyanskii and R. M. Macfarlane (Amsterdam: North Holland), 51, 1987.
4. J.S. Browder, S.S. Ballard, and P. Klocek, in *Handbook of Infrared Optical Materials*, ed. P. Klocek (Marcel Dekker, NY, 1991), p. 141.
5. Razeghi, M. *Optics & Photonics News* **6**, No. 8, page 16, (August, 1995).
6. Weber, M. J., ed., *CRC Handbook of Laser Science and Technology - Supplement 1: Lasers*, CRC Press, 1991.

Dr. Bradley Edwards

Dr. Edwards received his Ph.D. in physics from the University of Wisconsin at Madison. He is currently a staff scientist at Los Alamos National Laboratory working on space instrumentation. Dr. Edwards is a co-investigator on the ALEXIS satellite and involved in development of cryogenic detector systems at Los Alamos National Laboratory such as Ge gamma-ray, bolometer and superconducting tunnel junction detectors.