RADIATIVE COOLING

Harvesting the Coldness of the Universe

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The combination of the atmosphere's transparency in the 8-to-13-µm wavelength band and outer space's potential as a heat sink has opened up a new frontier in renewable-energy research.

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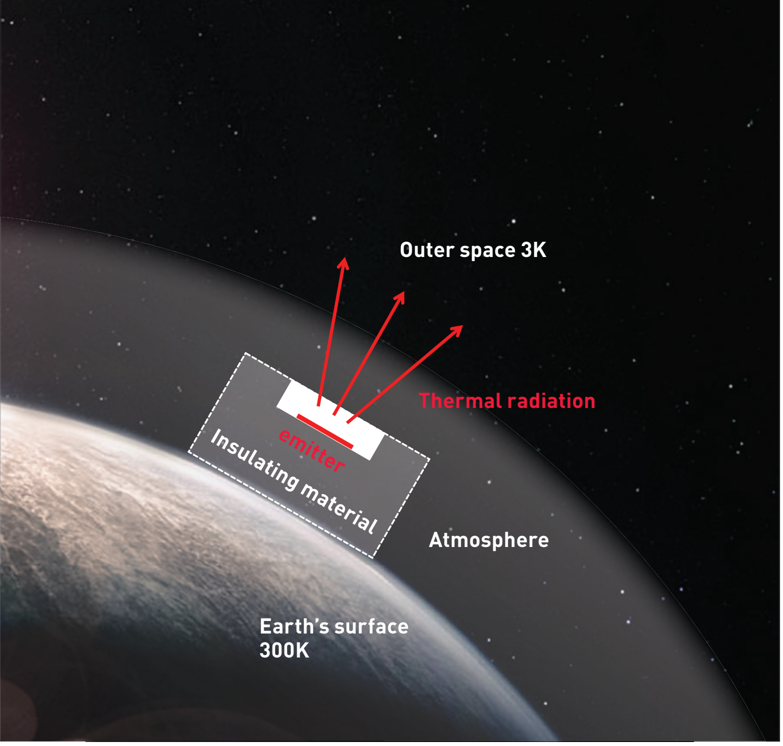


Under the second law of thermodynamics, efficient conversion of heat to work requires both a high-temperature heat source and a low-temperature heat sink. For humans, the sun, with a temperature of approximately 6000 K at the surface, represents the most important thermodynamic heat source, and harvesting solar energy has played a central role in the history of human civilization. Indeed, a significant portion of renewable-energy research today focuses on directly pulling energy from the sun—through such devices as solar thermal panels, which convert solar radiation to thermal energy, and photovoltaics, which convert it to electricity.

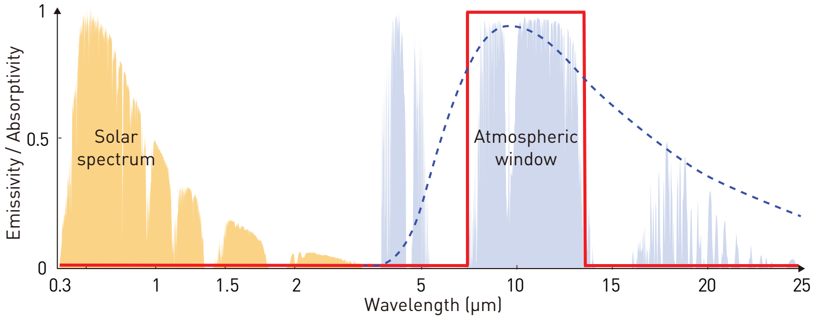
With all of this focus on the heat source, there has been far less attention to the heat-sink side. The vast majority of energy conversion processes at present simply use our ambient surroundings on Earth, at temperatures of roughly 300 K, as their thermodynamic heat sink. But the universe at large, with an average temperature of only around 3 K, offers a much better option. The opportunity to harness this coldness of the universe has broad implications for energy technology, and constitutes an important emerging frontier in renewable-energy research.

The ability to exploit the coldness of the universe comes through the technique of radiative cooling. In this feature, we look at work over the past five years, in our lab and others, that has moved this technique from the realm of speculation to tangible reality.

The basics of radiative cooling

Radiative cooling rests on a fact that's fundamental to determining Earth's temperature: the planet's atmosphere is highly transparent in the wavelength range of 8–13 μm. This transparency window has a very large overlap with the blackbody radiation spectrum at typical ambient temperatures near 300 K. Thus, in principle, any object on Earth, facing the sky, can radiate heat out to the universe through the atmospheric transparency window, and thus lower its temperature by remotely accessing the coldness of the universe.

More formally, radiative cooling can be understood as a simple heat balance, in which the net cooling power of the radiative cooler at a given temperature, $P_{cool}(T)$, is given by the power directly radiated out of the cooler, minus the power re-absorbed from incoming solar radiation and atmospheric thermal radiation, and lost through local conduction and convection. (For a more detailed treatment, see "The math of radiative cooling," p. 36.) Cooling occurs when $P_{cool}(T)$ is positive at the ambient temperature.



In radiative cooling, heat is emitted through the atmospheric transparency window between 8 and 13 μm—a prospect made possible, especially at night, by the broad spectral alignment between the transparency window and the peak in the radiation spectrum of a 300 K blackbody (dashed blue line). For daytime radiative cooling, the requirements are stricter: the materials involved must have near-zero absorptivity over the entire solar spectrum and also generate strong thermal radiation in the atmospheric transparency wavelength window (solid red line).

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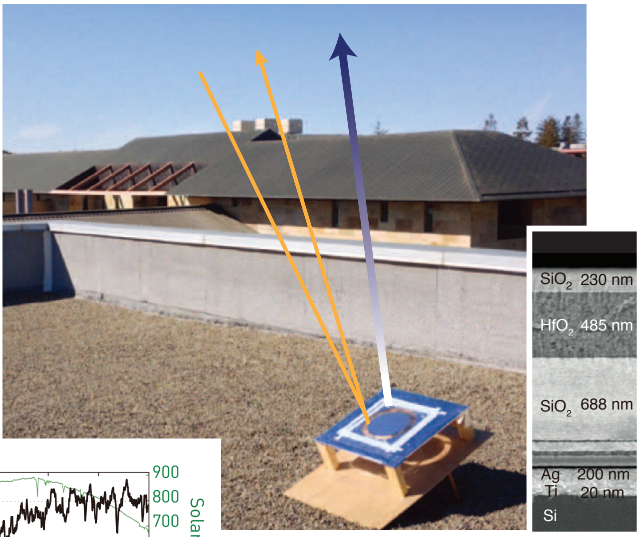
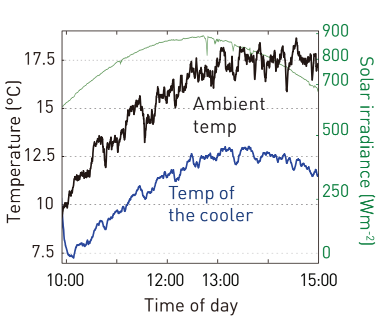
That condition is easily satisfied at night, when heat from the sun is absent. In fact, explorations of radiative cooling have a long history: in 400 B.C.E., well before the advent of electricity, people in the deserts of Iran were able to make and preserve ice in so-called Persian ice houses, or *yakhchāls*, at ambient temperatures above the freezing point of water. The cooling likely came from the combined effects of radiative and evaporative cooling. More recently, since the 1960s, nighttime radiative cooling has been demonstrated using materials such as plastics, TiO_2, SiO and MgO, as those materials have high emissivity in the mid-infrared wavelength range.

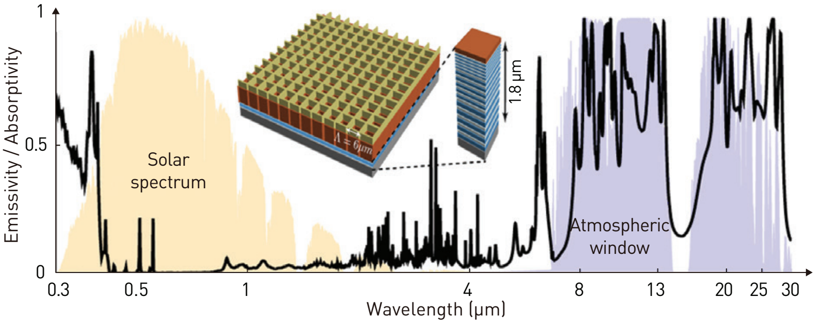


Cooling in the daytime

Radiative cooling during the day, when the need for cooling is usually greater, is even more desirable—and also more challenging. Because the sun heats up most materials during the day, achieving daytime radiative cooling requires a structure that has near-zero absorptivity over the entire solar spectrum and that also generates strong thermal radiation in the atmospheric transparency wavelength range of 8–13 μm. The combination has been difficult to achieve—and, as a result, while nighttime radiative cooling has a history stretching back millennia, daytime radiative cooling was first demonstrated only in 2014.

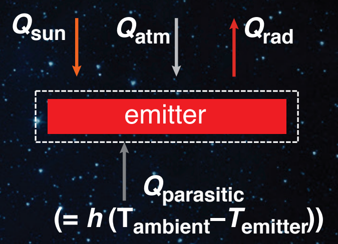


Those initial demonstrations rested on the realization that nanophotonic structures can be engineered to achieve the required broadband control of emissivity. In a 2013 study in our lab, Eden Rephaeli and colleagues reported the first theoretical design of a photonic structure capable of achieving daytime radiative cooling. The design consisted of two thermally emitting photonic crystal layers of SiC and quartz, underlain by a



The first nanophotonic design (chart above) and demonstration (photo and charts, upper right) of daytime radiative cooling. In the demonstration, the radiative cooler had a strong solar reflection and selective thermal radiation in the atmospheric-transparency window. Inset: Cross-sectional scanning electron microscope image of the radiative cooler.

E. Rephaeli et al. Nano Lett. **13**, 1457 (2013) / A.P. Raman et al. Nature **515**, 540 (2014)



The net cooling power is determined by the outgoing heat flux from the emission of the radiative cooler (Q_{rad}) and the incoming fluxes from the sun and atmosphere, as well as non-radiative heat transfer from conduction and convection.

The math of radiative cooling

To understand the radiative-cooling effect, consider a cooler of area A and temperature T, whose spectral and angular emissivity is $\epsilon(\lambda, \theta)$. When exposed to sun and sky, the cooler is subject to both solar irradiance and atmospheric thermal radiation, corresponding to the ambient air temperature, T_{amb}. The net cooling power, P_{cool}, of such a radiative cooler is given by:

$$P_{cool}(T) = P_{rad}(T) - P_{atm}(T_{amb}) - P_{sun} - P_{cond+conv}$$

Looking in more detail at the terms of this equation, the power radiated out by the radiative cooler, $P_{rad}(T)$, is

$$P_{rad}(T) = A \int d\Omega \cos\theta \int_0^\infty d\lambda \, I_{BB}(T, \lambda) \, \epsilon(\lambda, \theta)$$

where $\int d\Omega$ is the angular integral over a hemisphere, and $I_{BB}(T, \lambda)$ is the spectral radiance of a blackbody at temperature T and wavelength λ.

$$P_{atm}(T_{amb}) = A \int d\Omega \cos\theta \int_0^\infty d\lambda \, I_{BB}(T_{amb}, \lambda) \, \epsilon_{atm}(\lambda, \theta)$$

is the absorbed power due to downward atmospheric thermal radiation. The angle-dependent emissivity of the atmosphere is given by $\epsilon_{atm}(\lambda, \theta) = 1 - t(\lambda)^{1/\cos\theta}$, where $t(\lambda)$ is the atmospheric transmittance in the zenith direction. The incident solar power absorbed by the radiative cooler, P_{sun}, is

$$P_{sun} = A \int d\Omega \cos\theta \int_0^\infty d\lambda \, \epsilon(\lambda, \theta_{sun}) I_{AM1.5}(\lambda)$$

where the solar illumination is represented by $I_{AM1.5}(\lambda)$, the AM1.5 spectrum. Finally,

$$P_{cond+conv}(T, T_{amb}) = A h_c (T_{amb} - T)$$

gives the power lost due to convection and conduction. $h_c = h_{cond} + h_{conv}$ is a combined non-radiative heat coefficient that captures the collective effect of conductive and convective heating owing to the radiative cooler's local contact with external surfaces and adjacent air.

Radiative cooling takes place when the power radiated out of the cooler, $P_{rad}(T)$, is greater than the combined effects of the incoming sources of heat from sun, atmosphere, and local conduction/convection.

E. Rephaeli et al., Nano Lett. 13, 1457 (2013); A.P. Raman et al., Nature **515**, 540 (2014)

broadband solar reflector including three sets of five bilayers made of MgF_2 and TiO_2 with varying periods on a silver substrate. Theoretical modeling predicted that, under realistic atmospheric conditions and direct sunlight, this structure could passively achieve a temperature 40 to 60 °C below ambient temperature, or a cooling power density exceeding 100 W m^{-2}, at ambient temperatures of 300 K.

Following up on this theoretical work, Aaswath Raman and colleagues reported the first experimental demonstration of daytime radiative cooling in 2014. The multi-layer photonic structure the team developed, which consisted of seven dielectric layers deposited on top of a silver mirror, showed both a strong reflection for entire solar spectrum and a strong selective thermal emission in the atmospheric-transparency window.

The dielectric layers in particular were designed via a systematic optimization process that took into account real-world fabrication constraints. The top three layers, made of SiO_2 and HfO_2 with thicknesses on the order of hundreds of nanometers, are responsible for generating strong thermal radiation; the bottom four layers, of SiO_2 and HfO_2 and thicknesses on the order of tens of nanometers, serve to enhance the reflectivity of the silver mirror, especially in UV wavelengths. The device, when placed in a rooftop measurement setup, was able to reach a temperature that is 5 °C below the ambient air temperature—in spite of having about 900 W m^{-2} of sunlight directly impinging upon it.

To efficiently utilize radiative cooling, and reach the theoretically modeled temperature reductions, the photonic design needs to be combined with thermal considerations to minimize the parasitic heat gain of the cooler. In a 2016 experiment in our lab, Zhen Chen and colleagues were able to demonstrate a temperature reduction of more than 40 °C from ambient air temperature with a selective thermal emitter, by integrating the radiative cooler with a vacuum system that eliminated most parasitic heat gain.

Materials for daytime radiative cooling

Since this initial work in our lab at Stanford University, a variety of research groups have looked at material systems and structures for daytime radiative cooling. In 2017, the groups of Xiaobo Yin and Ronggui Yang at the University of Colorado reported a randomized glass–polymer hybrid, potentially manufacturable at a large scale, for daytime radiative cooling. The team embedded resonant polar dielectric microspheres randomly in a polymeric matrix, resulting in a material

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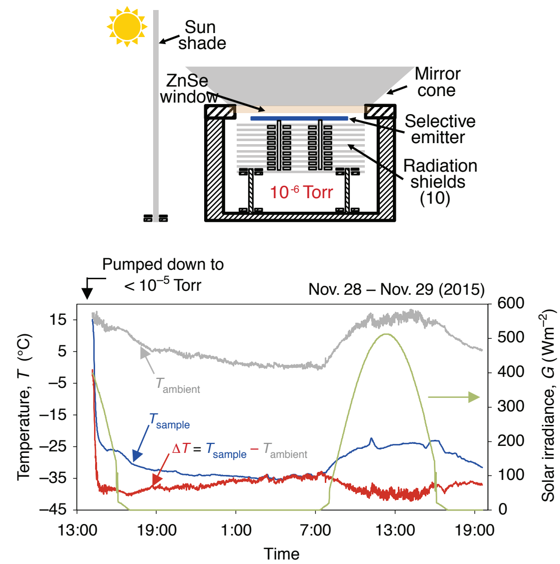


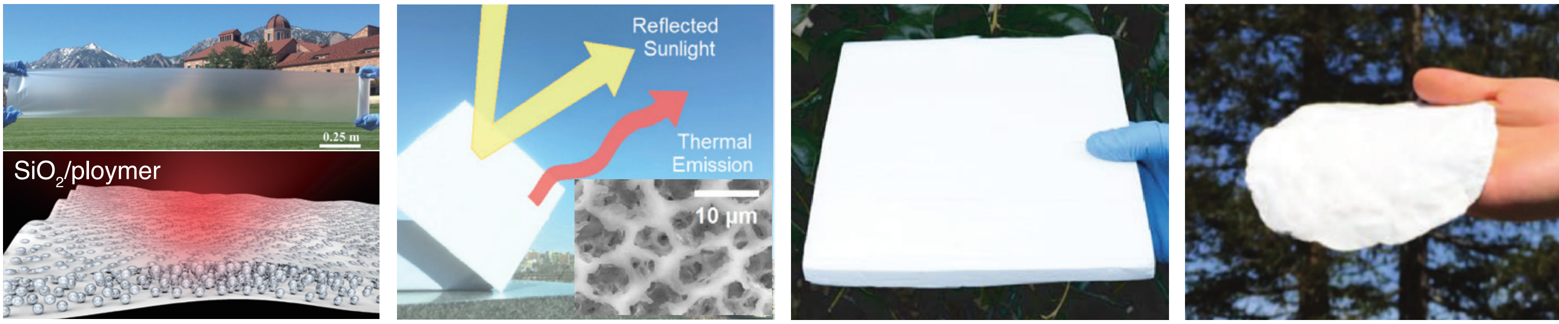
A vacuum system (top) achieved radiative cooling to deep sub-freezing temperatures by minimizing non-radiative heat loss (bottom). Chen et al. Nat. Commun. **7**, 13729 (2016)

that is fully transparent to the solar spectrum while having an infrared emissivity greater than 0.93 across the atmospheric window. When backed with a silver coating, this material showed a noontime radiative cooling power of 93 W m^{-2} under direct sunshine.

In 2018, the groups of Yuan Yang and Nanfang Yu at Columbia University developed a simple, inexpensive and scalable phase inversion–based method for fabricating hierarchically porous polymer coatings with good cooling performance. High, substrate-independent hemispherical solar reflectance (0.96 ± 0.03) and longwave infrared emittances (0.97 ± 0.02) allow for temperature drops of approximately 6 °C below ambient temperatures and cooling powers of around 96 W m^{-2} under solar intensities ranging from 750 to 890 W m^{-2}. Interestingly, the materials offer a paint-like simplicity of application—offering the prospect of large-scale radiative-cooling coatings.

Still other materials have broadened the application options for radiative cooling. Earlier this year, the groups of Liangbing Hu at the University of Maryland and Xiaobo Yin at the University of Colorado reported that, through a process of complete delignification and densification of wood, they had developed a wood-based material for radiative cooling, with a mechanical strength of 404.3 megapascals—more than eight times that of natural wood—that would make it suitable for structural applications. The cellulose nanofibers in this engineered material backscatter solar radiation and emit strongly in mid-infrared wavelengths, resulting in continuous cooling below ambient temperatures during both day and night.

Radiative-cooling materials have even been developed for personal thermal management of the human body. The research team of Yi Cui at Stanford



Recent advances in development of daytime radiative-cooling materials. Left to right: A randomized glass-polymer hybrid material for daytime radiative cooling; hierarchically porous polymer coatings for daytime radiative cooling; a radiative-cooling structural material; and an outdoor radiative-cooling textile.

Zhai et al., Science **355**, 1062 (2017) / J. Mandal et al., Science **362**, 315 (2018) / Li et al., Science **364**, 760 (2019) / Cai et al., Adv. Mater. **30**, 1802152 (2018)

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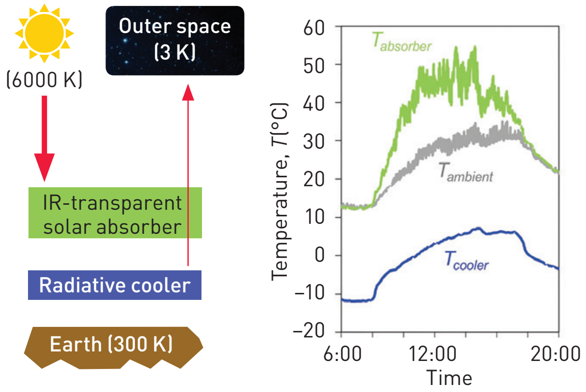

Simultaneous energy harvesting from the sun and outer space.
Chen et al., Joule **3**, 101 (2019)

University, for example, developed a novel, spectrally selective nanocomposite textile for radiative outdoor cooling, using polyethylene embedded with zinc oxide nanoparticles. In experiments under peak daylight conditions, this textile, by reflecting more than 90% of the solar irradiance and selectively transmitting out the body's thermal radiation, allowed simulated human skin to avoid overheating by 5–13 °C compared with conventional textiles like cotton.

Application opportunities

As the discussion above suggests, the application space for radiative cooling is potentially wide-ranging. In the rest of this feature, we look at a number of application opportunities, including several examples from our lab.

One obvious application area is in air-conditioning systems for buildings, which requires that radiative cooling be integrated into the larger building air-conditioning systems to deliver cooling where it's needed. To this end, Eli Goldstein and coworkers demonstrated large-area (greater than 1 m^2) panels that can passively, non-evaporatively cool fluids, such as water, to 3–5 °C below ambient temperatures at flow rates relevant to air-conditioning systems. Feeding such cooled water into the condenser of a vapor-compression system can boost the overall efficiency of the cooling system by 20% or more.

Radiative cooling also offers a unique tool for harvesting water from atmospheric humidity. Cooling to a temperature below the dew point condenses water on the radiative cooler, which is collected to produce clean water. This mechanism is already utilized by commercial radiative dew condensers—but thus far these have worked only at night. To extend these systems into the daytime hours, the groups of Zongfu Yu at University of Wisconsin–Madison and Qiaoqiang Gan at the State University of New York at Buffalo have demonstrated a daytime radiative condenser that reflects almost all solar radiation, and can thus condense atmospheric water even in direct sunlight, potentially doubling the production of purified water over a 24-hour period.

Still other opportunities lie in combining radiative cooling with more conventional forms of solar energy harvesting. For instance, the heating of photovoltaic solar cells can diminish both their power conversion efficiency and their useful life; several recent demonstrations have shown how radiative cooling could be used to overcome those disadvantages by reducing the operating temperature of solar cells. Another recent experiment showed how solar energy harvesting and radiative cooling could act in tandem, by placing a solar thermal absorber that is transparent in mid-infrared above a radiative cooler. The solar absorber is heated to 24 °C above the ambient temperature, and provides a shading mechanism that enables the radiative cooler to reach 29 °C below the ambient temperature.

Future directions

The recent advances in radiative cooling build upon the developments in nanophotonics for the control of thermal radiation, and have emerged as an important direction for renewable-energy research. Future work is likely only to further improve the technique's performance and its integration into existing cooling systems, as well as spurring new application areas. We close this

The development of radiative cooling should lead to many new opportunities. The result will be a strong, positive impact on energy, environment and the sustainability of our society.

broad overview by pointing out a few emerging directions in radiative cooling.

From a basic thermodynamic perspective, any temperature gradient can be used to generate power, and several research groups have explored the theoretical limits of using the temperature difference between the Earth and outer space as a power source in its own right. Two recent experiments in our lab showed that a semiconductor photodiode at ambient temperature can directly generate power when the diode faces the sky, as well as nighttime power generation through radiative cooling using a thermoelectric device. Future efforts may significantly improve the efficiency of such power-harvesting systems.

Most existing radiative-cooling systems are static, in that their thermal emissivity is fixed when they are constructed. As temperature varies across days and seasons, however, and as environmental temperature will sometimes drop below a threshold where cooling is no longer desired—for example, during nighttime in winter—static radiative cooling may no longer be desirable, and may even increase the energy consumption if heating is required to compensate. Therefore, for many applications, an active radiative-cooling system that can enable or disable cooling depending on the need would be highly desirable. One possibility, recently explored theoretically in our group, would be a tunable radiative-cooling system based on phase-change materials.

Making radiative cooling practical also requires that the materials in these systems be reliable in outdoor environments. Interestingly, there's a strong correlation here between direct solar energy harvesting and radiative cooling—radiative cooling typically works best in environments where solar energy harvesting is most effective. As a result, recent technology advances in the reliability of solar-energy-harvesting systems can potentially be used in developing radiative-cooling systems.

In light of recent fundamental advances and practical applications, the development of radiative cooling should lead to many new opportunities. The result will be a strong, positive impact on energy, environment and the sustainability of our society. OPN

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For an expanded list of references, go online: www.osa-opn.org/link/radiative-cooling.