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Color-preserving daytime radiative cooling

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We introduce a general approach to radiatively lower the temperature of a structure, while preserving its color under sunlight. The cooling effect persists in the presence of considerable convective and conductive heat exchange and for different solar absorptances. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4835995]

The color of an object under sunlight is typically determined by its absorption spectrum over solar wavelengths. This absorption of sunlight in turn will cause the object to heat up to temperatures substantially above ambient air temperatures. Here, we ask a general question: is there an approach that allows one to lower the temperature of the object under solar illumination, while preserving its color? In this Letter, we tackle this challenge by presenting a general strategy for color-preserving daytime radiative cooling.

The Earth's atmosphere has a transparency window for electromagnetic waves between 8 and 13 μm that coincides with peak thermal radiation wavelengths at terrestrial temperatures. For a given object, creating an imbalance between the heat radiated outwards through the transparency window, and the absorbed radiation from the environment, is key to achieving radiative cooling. For nighttime cooling, the absorbed radiation results from the thermal emission of the atmosphere. There have been numerous studies on radiatively cooling an object below ambient temperatures during the night.[1–10] For daytime cooling, one must also take into account solar absorption. In order to achieve an equilibrium temperature below the ambient in daytime radiative cooling,[1,11–13] it is crucial to achieve over 88% solar radiation reflection.[11] But covering an object with a strong solar reflector of course significantly alters its color. In contrast to all previous works on daytime radiative cooling,[1,11–13] here we do not seek to control the object's solar radiation reflection as it is constrained by the object's color, which we seek to preserve. And hence, we do not seek to achieve an equilibrium temperature that is below the ambient. Instead, for an object with a given color, and thus a given amount of solar absorption, our objective is to lower its temperature as much as possible, while preserving its color. Achieving such a color-preserving daytime radiative cooling effect is important because it enables energy-free, passive cooling in situations where the color of an object needs to be maintained for aesthetic or functional purposes.

Before we discuss our design, we briefly review the procedure of our analysis. We consider a structure at temperature T, with a spectral and angular emissivity $\epsilon(\lambda,\Omega)$. The structure is exposed to a clear sky and is subject to solar irradiance and atmospheric irradiance corresponding to an ambient temperature T_{amb}. The net cooling power per unit area of a structure, $P_{net}(T)$, is given by

$$P_{net}(T) = P_{rad}(T) - P_{atm}(T_{amb}) - P_{sun}, \tag{1}$$

where

$$P_{rad}(T) = \int d\Omega \cos\theta \int_0^\infty d\lambda I_{BB}(T,\lambda)\epsilon(\lambda,\Omega) \tag{2}$$

is the power radiated by the structure per unit area,

$$P_{atm}(T_{atm}) = \int d\Omega \cos\theta \int_0^\infty d\lambda I_{BB}(T_{amb},\lambda)\epsilon(\lambda,\Omega)\epsilon_{atm}(\lambda,\Omega) \tag{3}$$

is the absorbed power per unit area emanating from the atmosphere, and

$$P_{sun} = \int_0^\infty d\lambda \epsilon(\lambda,0) I_{AM1.5}(\lambda) \tag{4}$$

is the incident solar power absorbed by the structure per unit area. Here, $\int d\Omega = \int_0^{\pi/2} d\theta \sin\theta \int_0^{2\pi} d\phi$ is the angular integral over a hemisphere. $I_{BB}(T,\lambda) = (2hc^2/\lambda^5)/[e^{hc/(\lambda k_B T)} - 1]$ is the spectral radiance of a blackbody at temperature T, where h, c, k_B, and λ, are the Planck constant, the velocity of light, the Boltzmann constant, and wavelength, respectively. In obtaining Eqs. (3) and (4), we used Kirchhoff's law to replace the structure's absorptivity with its emissivity $\epsilon(\lambda,\Omega)$. The angle-dependent emissivity of the atmosphere is given by[3] $\epsilon_{atm}(\lambda,\Omega) = 1 - t(\lambda)^{1/\cos\theta}$, where $t(\lambda)$ is the atmospheric transmittance in the zenith direction.[14] In Eq. (4), the solar illumination is represented by $AM1.5$ Global Tilt spectrum with an irradiance of 964 W/m^2, which represents the average solar conditions of the continental U.S. We assume that the structure is facing the sun. Hence, the term P_{sun} is devoid of an angular integral, and the structure's emissivity is represented by its value in the zenith direction, $\theta = 0$.

We now introduce our design principle for color-preserving daytime radiative cooling. We start by presenting an idealized design to highlight some of the design considerations, and examine the theoretical potential for this principle's performance. We consider an "original" structure having an emissivity/absorptivity spectrum as shown in Fig. 1(a), where

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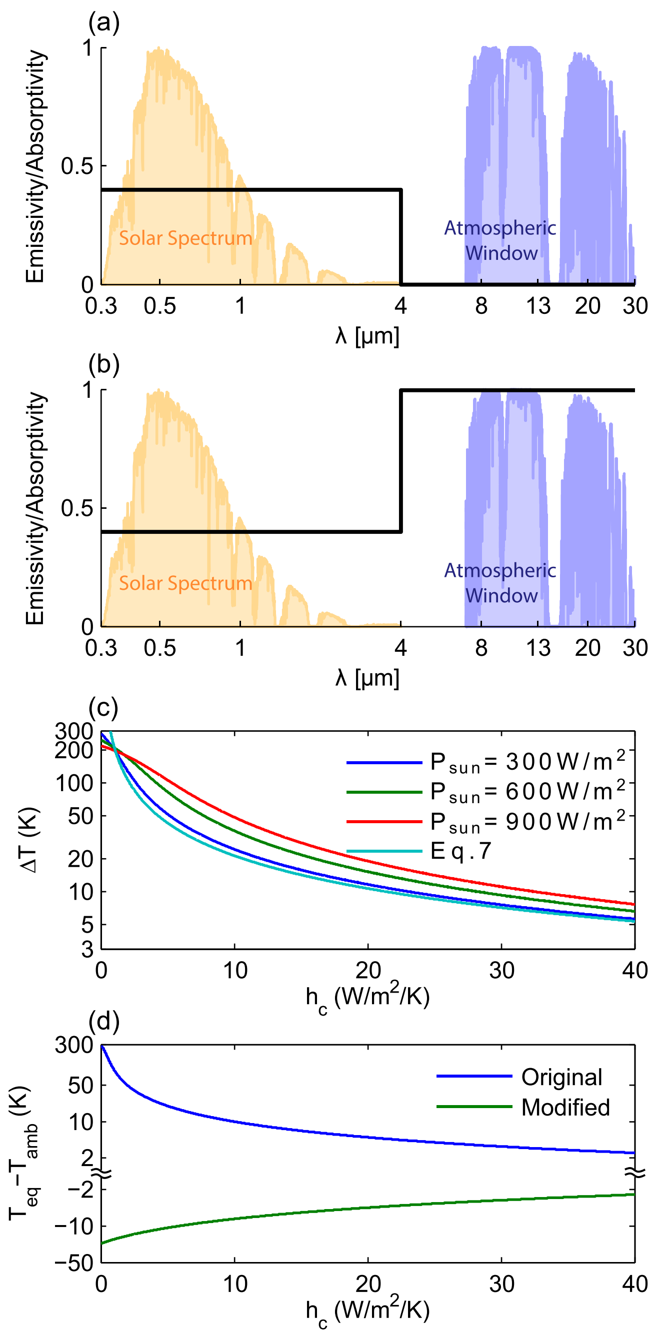

FIG. 1. (a) The black curve is the emissivity/absorptivity spectrum the original structure which, for simplicity, absorbs sunlight at a flat value and emits no thermal radiation. The normalized solar radiation spectrum of $AM1.5$ spectrum is shown in yellow. The normalized atmosphere transmission spectrum is shown in magenta. (b) The black curve is the ideal emissivity/absorptivity spectrum of a modified structure, which has unity emissivity at thermal wavelengths. (c) The difference between the equilibrium temperatures for the original and the modified structures ($\Delta T = T_{eq,original} - T_{eq,modified}$), at $T_{amb} = 300$ K, as a function of non-radiative heat exchange coefficient h_c, for different amounts of solar absorption P_{sun}. (d) $T_{eq} - T_{amb}$ as a function of h_c, for $P_{sun} = 100$ W/m^2, at $T_{amb} = 300$ K.

for simplicity, the absorptivity at solar wavelengths is assumed to be uniformly flat. We also assume that this original structure has zero emissivity at thermal radiation wavelengths. Thus, this original structure represents a worst case scenario as far as radiative cooling is concerned since it does not emit any thermal radiation. To preserve the color of the original structure, while lowering its temperature via radiative cooling, our design principle is to create a "modified" structure that maintains the original structure's absorptivity at solar wavelengths, while being highly emissive at thermal wavelengths. We note that, generally, $(P_{rad} - P_{atm})$ does not dominate the solar absorption, and for $T = T_{amb}$, $P_{net} < 0$, implying that the structure heats up even with radiative cooling, reaching an equilibrium temperature $T > T_{amb}$. However, with the large $(P_{rad} - P_{atm})$ added by the radiative cooling, the modified structure can reach thermal equilibrium at a lower temperature than the original structure. An ideal design to maximize cooling capability has an emissivity/absorptivity spectrum as in Fig. 1(b), where the absorptivity is unchanged at solar wavelengths to preserve the color, while emissivity is unity at thermal wavelengths. This ideal design is derived by maximizing $(P_{rad} - P_{atm})$, when $T > T_{amb}$. We note the ideal emissivity spectrum here is different from considerations in nighttime[1–10] or daytime[1,11–13] radiative cooling, where achieving high emissivity inside, while negligible emissivity outside, the atmospheric window, is crucial for cooling to the lowest possible temperature below the ambient.

From the emissivity/absorptivity spectrum of the ideal design (Fig. 1(b)), at $T = T_{amb} = 300\,\mathrm{K}$, $P_{rad}(T) = 424$ W/m^2, $P_{atm}(T_{amb}) = 209\,\mathrm{W/m^2}$, and $P_{rad} - P_{atm} = 215$ W/m^2. The large $(P_{rad} - P_{atm})$, though smaller than solar absorption P_{sun}, will nevertheless cool the structure to a lower temperature compared with the original structure.

We now demonstrate that our general approach can lower the temperature of the structure considerably, even in the presence of large non-radiative heat exchange. The effects of conductive and/or convective heat exchange are accounted for by adding a term $P_{cond+conv} = h_c(T - T_{amb})$ with $T > T_{amb}$ in Eq. (1). Thermal equilibrium temperature (T_{eq}) is then determined by

$$P_{rad}(T_{eq}) - P_{atm}(T_{amb}) - P_{sun} + h_c(T_{eq} - T_{amb}) = 0. \quad (5)$$

The presence of 1 m/s, 3 m/s, and 12 m/s wind speeds would result in a combined non-radiative heat coefficient value h_c of approximately 6, 12, and 40 W/m^2/K, respectively.[12] In Fig. 1(c), we show the dependence of the equilibrium temperature difference $(\Delta T = T_{eq,original} - T_{eq,modified})$ between the original structure and the modified structure on h_c, for different P_{sun}. The modified structure achieves considerably lower equilibrium temperatures than the original structure across a large range of h_c. Even with the presence of non-radiative heat exchange coefficient as large as $h_c = 40\,\mathrm{W/m^2/K}$, the temperature reduction is a meaningful 7.7 K for 900 W/m^2 incident solar absorption (a value that corresponds to the sun's irradiance at mid-day).

Our general approach of color-preserving radiative cooling is also robust against different values of solar absorption (P_{sun}). We first apply Eq. (5) both for the original and modified structures. Exploiting the fact that for the original structure $P_{atm}(T_{amb}) \approx 0$, $P_{rad}(T) \approx 0$ (when T is not exceedingly high so that thermal emission at solar wavelengths is negligible), and that solar absorption is the same between two structures, we have

$$\Delta T = T_{eq,original} - T_{eq,modified} \approx \frac{P_{rad,modified}(T_{eq,modified}) - P_{atm,modified}(T_{amb})}{h_c}. \quad (6)$$

As $P_{rad,modified}(T_{eq,modified})$ increases as $T_{eq,modified}$, and $T_{eq,modified} > T_{amb}$, from Eq. (6), we have

$$\Delta T \geq \frac{P_{rad,modified}(T_{amb}) - P_{atm,modified}(T_{amb})}{h_c}. \quad (7)$$

Equation (7) becomes an equality when heating is negligible, i.e., $T_{eq,modified} \approx T_{amb}$ (for large h_c and small P_{sun}). We note that Eq. (7) is independent of the absorptivity at solar wavelengths, as at typical terrestrial ambient temperatures $T = T_{amb}$; the corresponding thermal radiation spectrum does not extend into solar wavelengths. Thus, Eq. (7) provides a lower bound on the achievable temperature reduction. When $T_{amb} = 300\,\mathrm{K}$, Eq. (7) amounts to $\Delta T \geq \frac{215\,\mathrm{W/m^2}}{h_c}$, which is a general lower bound for ΔT between the original structure and the modified structure with ideal design and independent of solar absorption. Therefore, at $T_{amb} = 300\,\mathrm{K}$, the temperature reduction is at least 18 K for $h_c = 12\,\mathrm{W/m^2/K}$ (corresponding to 3 m/s wind speed) and 5.4 K for $h_c = 40\,\mathrm{W/m^2/K}$ (corresponding to 12 m/s wind speed). From Fig. 1(c), we see the lower bound for temperature reduction provided by Eq. (7) is consistent with actual calculations for different P_{sun}. Only in the regime of $h_c < 1\,\mathrm{W/m^2/K}$, which is far from most practical situations, do we see a violation of Eq. (7). In such a case, the heating of the original structure is so strong that the thermal radiation partially extends into solar wavelengths, making P_{rad} non-negligible for the original structure. Equation (7) therefore provides a convenient way to estimate the achievable temperature reduction for practical scenarios.

We also show the equilibrium temperatures of the original and modified structures, for $P_{sun} = 100\,\mathrm{W/m^2}$ in Fig. 1(d). Here, the P_{sun} is sufficiently small, such that the modified structure cools down below ambient, while the original structure still heats up substantially.

We now present and analyze a specific design to achieve color-preserving daytime radiative cooling. Silicon is used as our example material for creating color as it has a very small extinction coefficient over thermal wavelengths at typical temperatures,[15] while silicon nanostructures can be designed to exhibit various colors by their geometric properties.[16] As an example, the original structure we study consists of a periodic array of silicon nanowires on top of an aluminum substrate (Fig. 2(a)). The silicon nanowires have a square cross section with 100 nm side length, and the periodicity between the nanowires is 500 nm. Using the rigorous coupled-wave analysis (RCWA) method,[17] we perform a calculation of the absorptivity over an ultra broadband wavelength range between 0.3 and 30 μm with a wavelength step-size of 2 nm in Fig. 2(b). The absorptivity shown here is the averaged absorptivity for both polarizations at normal incidence. We see from Fig. 2(b) that the emissivity of the original structure at thermal wavelengths is negligible, while there is substantial solar absorption present at optical wavelengths. The *CIE* 1931 chromaticity coordinates[18] of the original structure for normal incidence reflection, and averaged over two polarizations, are calculated to be $(x = 0.3598, y = 0.3342)$, which corresponds to a light pink color (Fig. 2(a)).

To preserve the color of the silicon nanostructure, while lowering its temperature via radiative cooling, we seek to

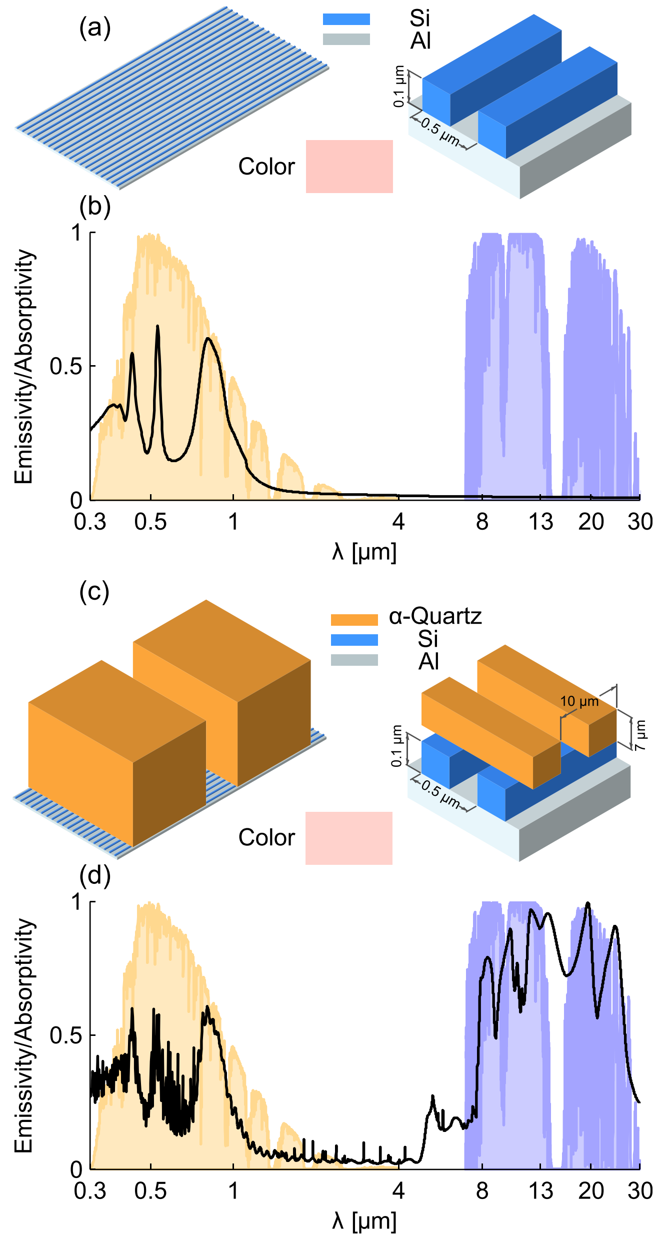

FIG. 2. (a) Schematic of the original structure, with silicon nanowire array on top of an aluminum substrate. A disproportionate schematic is also shown to clarify the geometry. The color for reflection at normal incidence under sunlight is shown. (b) The black curve is the emissivity/absorptivity spectrum of the original structure. The normalized solar radiation spectrum of the *AM*1.5 spectrum is shown in yellow. The normalized atmosphere transmission spectrum is shown in magenta. (c) Schematic of the modified structure, with quartz bar array on top of the original structure of silicon nanowires. (d) The black curve is the emissivity/absorptivity spectrum of the modified structure.

identify and place atop the silicon a material that is transparent at solar wavelengths, while strongly emissive in the atmospheric transparency window between 8 and 13 μm. α-Quartz is one such material: it has two phonon-polariton resonances at the relevant thermal wavelengths and is transparent over visible wavelengths. To achieve color-preserving daytime radiative cooling, we consider the design shown in Fig. 2(c), where a periodic array of quartz bars sits on top of the original structure. Each quartz bar has a square cross section with 7 μm side length, and the periodicity between quartz bars is 10 μm. The quartz bars and silicon nanowires are orthogonal to each other. We show the modified structure's absorptivity over the same wavelength range of 0.3–30 μm in Fig. 2(d).

With the quartz array, the modified structure has large emissivity at thermal wavelengths, while the absorption spectrum in the visible range is almost entirely unchanged compared with the original structure. The *CIE* 1931 chromaticity coordinates are calculated to be $(x = 0.3524,\ y = 0.3362)$, corresponding as before to light pink (Fig. 2(c)). Therefore, adding the quartz bar array on top of the original structure maintains the original color.

For the modified structure with quartz bars, at $T = T_{amb} = 300\,\mathrm{K}$, $P_{rad}(T) = 273.3\,\mathrm{W/m^2}$, $P_{atm}(T_{amb}) = 117.7\ \mathrm{W/m^2}$, and $P_{rad} - P_{atm} = 155.6\,\mathrm{W/m^2}$. For the original structure, at $T = T_{amb} = 300\,\mathrm{K}$, $P_{rad}(T) = 5.5\,\mathrm{W/m^2}$, $P_{atm}(T_{amb}) = 2.7\,\mathrm{W/m^2}$, and $P_{rad} - P_{atm} = 2.8\,\mathrm{W/m^2}$. On the other hand, $P_{sun} = 274.0\,\mathrm{W/m^2}$ for the modified structure with quartz bars, while for the original structure $P_{sun} = 268.9\,\mathrm{W/m^2}$. Hence, the solar absorption is nearly the same between the two structures, in consistency with our requirement that the two structures should have the same color. With the large $(P_{rad} - P_{atm})$ in the presence of the quartz bars, the modified structure reaches thermal equilibrium at a lower temperature than the original structure.

We show the dependence of equilibrium temperature (T_{eq}) on h_c for the modified structure with quartz bars, and the original structure, in Fig. 3(a). Under sunlight, both structures heat up considerably, but T_{eq} for the modified structure with quartz bars is considerably lower than the original structure (Fig. 3(a)). From Fig. 3(b), the temperature reduction enabled by the quartz bars is 31.4 K for $h_c = 6\,\mathrm{W/m^2/K}$ (corresponding to 1 m/s wind speed), 14.4 K for $h_c = 12\,\mathrm{W/m^2/K}$ (corresponding to 3 m/s wind speed), and 4 K for a non-radiative heat exchange coefficient as large as $h_c = 40\,\mathrm{W/m^2/K}$ (corresponding to 12 m/s wind speed). Therefore, the added quartz bars can lower the temperature of the structure by a meaningful amount under direct sunlight, even in the presence of considerable non-radiative heat exchange associated with wind or thermal contact.

We have assumed a uniform temperature distribution within the structures. In general, one can assume a uniform temperature distribution within a structure if its Biot number[19] $Bi \equiv hL/\kappa \ll 1$. In our structure, the heat transfer coefficient h is generally smaller than $40\,\mathrm{W/m^2/K}$, the characteristic length scale L is about 10 μm, and thermal conductivity κ is larger than 1 W/m/K. Therefore, the Biot number Bi is approximately $0.0004 \ll 1$.

We note that the use of silicon is not essential to the performance of this scheme, and silicon can be replaced by other materials. With this approach one can meaningfully lower the temperature of a structure, while preserving its color by utilizing a microstructure of optically transparent thermally emissive materials like quartz or SiO_2. We believe this general approach can be a powerful enabling tool for a broad array of applications where energy-free, passive cooling is in demand. One possible application may be outdoor or technical clothing, where one can reduce the temperature of the cloth in an entirely passive manner, while maintaining its original color for aesthetic or functional purposes. Similarly, this approach may also be useful to prevent or reduce the overheating of devices whose efficiency seriously degrades at higher temperature, such as outdoor electronic devices. More broadly, we believe this approach shows that radiative cooling through sky-access can be a useful and powerful tool even when the equilibrium temperatures achieved are above the ambient.

In conclusion, we have introduced a general approach to radiatively lower the temperature of a structure, while preserving its color. We have shown that such an approach works in the presence of substantial non-radiative heat exchange and is robust against all values of solar absorption that could heat the structure. Moreover, we have provided a concrete numerical implementation of this approach based on realistic three-dimensional simulations and real material parameters.

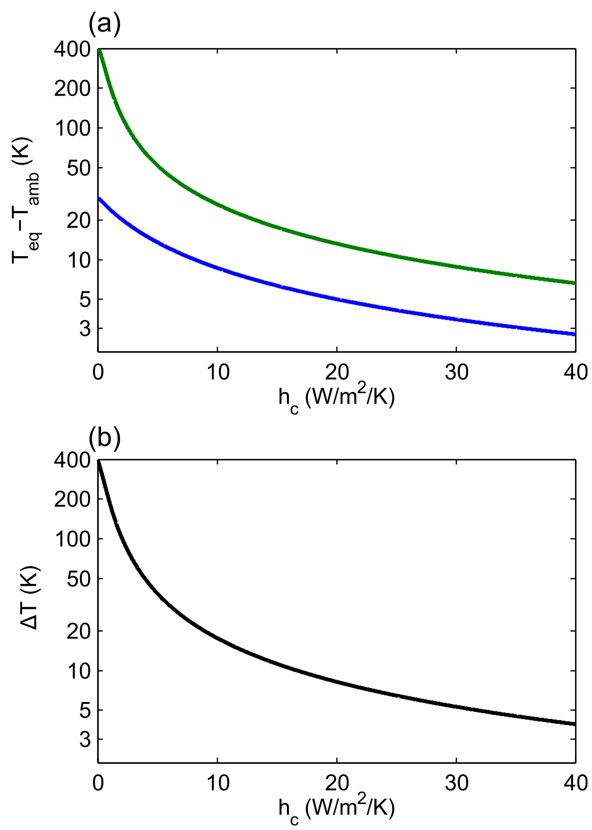

FIG. 3. (a) Equilibrium temperature (T_{eq}) subtracted by ambient temperature $T_{amb} = 300$ K, for the modified structure with quartz bar array (blue curve) and the original structure (green curve), as a function of non-radiative heat exchange coefficient h_c. (b) The difference (ΔT) between the equilibrium temperatures for the original and the modified structures, as a function of h_c. The modified structure has a meaningfully lower equilibrium temperature even for large values of h_c.

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