



Enhance radiative cooling performance of nanoparticle crystal via oxidation

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ABSTRACT

Nanoparticle-crystal is a promising candidate for large scale metamaterial fabrication. However, in radiative cooling application, the maximum blackbody radiation wavelength locates far from metal's plasmon wavelength. In this paper, it will be shown if the metallic nanoparticle crystal can be properly oxidized, the absorption performance within room temperature blackbody radiation spectrum can be improved. Magnetic polariton and surface plasmon polariton have been explained for the mechanism of absorption improvement. Three different oxidation patterns have been investigated in this paper, and the results show they share a similar enhancing mechanism.

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1. Introduction

With the well-designed subwavelength structure, metamaterial can manipulate the propagation and redistribution of light in sub-wavelength scale [1]. As originally employed to generate negative index of refraction [2], metamaterial has been gradually utilized in energy harvesting and thermal radiation management [3–5]. Most recently, its application on radiative cooling has drawn a lot of attention. With properly designed structure, metamaterial can emit thermal electromagnetic wave in specific spectrum with high intensity. When this high emission band matches with the transparency window in the atmosphere between 8 to 13 μm , the device which is exposed to the sky can radiate heat to outer space and cool itself down. This technique is well known as radiative cooling, which can maintain a temperature below the ambient air, even at direct solar irradiation [6]. As it can cool the subject without any electricity input, radiative cooling gains various applications, like solar cell [7], high power LED [8], and textile that can be dressed [9].

To build a radiative cooling device has gradually became truth [10]. But one major obstacle is the selective surface should be fabricated in a large scale, which is quite demanding for conventional fabrication method. In this way, novel strategies have been proposed to produce metamaterial and handle this problem. For instance, glass-polymer hybrid metamaterial has been proposed [11]. With the unique coupling between microsphere and light, the

emission band of SiO_2 has been greatly expanded. Other methods toward large-scale metamaterials have also been reported, like multilayer structure [6,10] and nanoporous textile [9]. Besides those efforts, nanoparticle crystal has also gained a lot of attention. Nanoparticle crystal refers to metasurface in which nanoparticles are closely packed in a similar pattern with atomic crystal [12–14]. The prominent merit of such kind of metamaterial is the little time required for fabrication, comparing with conventional method likes lithography. Nanoparticles can be produced in large scale via chemical synthesis, and when they are transferred into periodic pattern, resonance can be excited and results in a highly selective absorptivity [15].

For radiative cooling device, even the goal to enhance absorption/emission is similar with solar harvesting, many well-studied solar harvesting techniques cannot be applied on it. That's because radiative cooling device interests mid-infrared photon, whose energy is much lower than visible photon that solar harvesting focuses. For instance, the intrinsic localized surface plasmon mode for Au nanoparticles locates at around 530 nm [16], leaving mere chance to apply this emission mode on radiative cooling. Thus, to enhance the emission performance, resonance should be excited. But back to the fabrication, to excite resonant emission, geometric features need to be well-controlled. Dielectric layer should be properly placed among metallic structures to support resonant modes [17,18].

In some structure made of abundant material like Al, oxidation of metal can easily occur, which results in a natural dielectric layer. It has been shown that if nanoparticle can be properly oxidized, the emission performance of a nanoparticle-trimer can be obviously improved [19]. In this paper, it will be shown that if the metallic nanoparticle crystal can be properly oxidized, enhancement of

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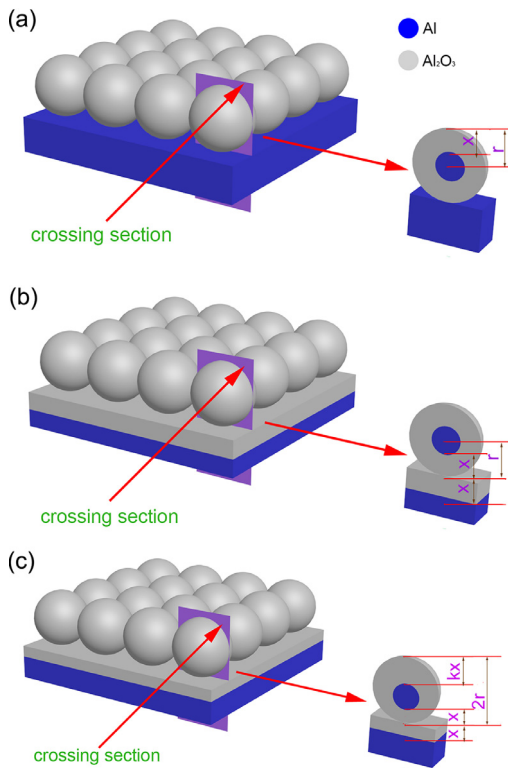


Fig. 1. Schematic of (a) PO pattern, (b) PSEO pattern, (c) PSUO pattern.

selective emission/absorption can be achieved. The stack pattern of nanoparticles can be cubic or hexagonal. As the optical responses for two kinds of crystals are similar [20], we merely focus on cubic nanoparticle crystal for simplicity reason. In our previous work [20], we have designed a double layer nanoparticle crystal with polarization independent absorption performance in near-infrared region, and explored its potential use in thermophotovoltaic. In this work, we focus the oxidation effect on monolayer nanoparticle crystal's mid-infrared emission spectrum, which radiative cooling device interests.

Three crystal oxidation patterns have been investigated in this paper, as particle-oxidized (PO), particle-substrate-evenly-oxidized (PSEO) and particle-substrate-unevenly-oxidized (PSUO) pattern. PO pattern occurs when metallic particles are oxidized before deposited onto the substrate. When the oxidation occurs after particles are deposited on the substrate, PSEO and PSUO patterns come into existence. Even the three oxidation patterns are different, it will be shown the mechanisms of emission enhancement are similar. The three patterns are illustrated in Fig. 1, in which the radius of particle r is set as $5 \mu\text{m}$, and period of monolayer nanoparticle crystal $P = 2r = 10 \mu\text{m}$ in this paper. Abundant metallic material Al is chosen for unoxidized particle and substrate. The optical models of Al and Al_2O_3 are obtained from Palik's book [21]. In PO pattern, shown as Fig. 1(a), the particles are oxidized while substrate is not. The thickness of Al_2O_3 layer inside the particle is x . In PSEO pattern, as Fig. 1(b), the particles and substrate are all oxidized, and the Al_2O_3 layers in particle and substrate are same in thickness as x . In PSUO pattern, illustrated as Fig. 1(c), oxidation occurs in both particle and substrate, but thicknesses of Al_2O_3 layers are different. In the side where particle contacts substrate, the thicknesses of Al_2O_3 layer are same in two subjects, both as x . On the other side of particle, the thickness of Al_2O_3 layer is kx , where k is the factor considering the oxidation non-uniform. In this paper, the oxidation rate (OxR) is defined as x/r . The PO pattern will be

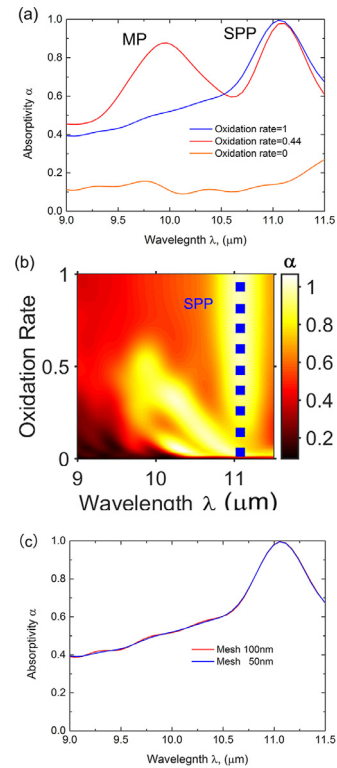


Fig. 2. (a) Absorptivity spectrum for PO nanoparticle crystal with OxR = 1, 0.44, 0. (b) Absorptivity of PO as a function of wavelength and OxR. The blue squares indicate resonant wavelength of SPP predicted by dispersion relationship. (c) Absorptivity spectrum for PO nanoparticle crystal with OxR = 0, calculated with different mesh, 100 nm square mesh and 50 nm square mesh.

comprehensively discussed and later we would show the mechanism obtained from PO pattern can be applied to PSEO and PSUO patterns.

2. Resonant absorption in PO pattern

2.1. Absorption spectrum for PO pattern

The absorptivity spectrum for PO pattern has been plotted in Fig. 2(a) with OxR = 1, 0.44, 0. It can be seen with OxR = 0, absorptivity stays in a low value in the whole spectrum, and no absorption peak can be observed. With the OxR increase to 0.44, double major absorption peaks located at $9.80 \mu\text{m}$ and $11.07 \mu\text{m}$ can be seen. They will be explained as magnetic-polariton (MP) and surface plasmon polariton (SPP). In the moderate oxidation case, 0.1 for example, there is a minor peak near MP. That peak also belongs to MP, and the field distribution is very similar with major MP case. For simplicity reason, we focus on the major MP. With the OxR increases to 1, MP mode disappears, while SPP changes very slightly in absorptivity.

The resonant wavelength of SPP can be predicted by its dispersion relationship [22]:

$$|k_{\text{spp}}| = \frac{\omega}{c_0} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \quad (1)$$

Surface plasmon polariton can be excited in the interface of two dissimilar materials with permittivities ε_1 and ε_2 . In the periodic grating, the in-plane wavevector can be matched for diffracted wave with grating Bloch condition:

$$k_{||} = \left(k_{x,\text{inc}} + \frac{2\pi m}{P_x} \right) \hat{x} + \left(k_{y,\text{inc}} + \frac{2\pi n}{P_y} \right) \hat{y} \quad (2)$$

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