

1 Article

2 Extraordinary light trapping enhancement in silicon 3 solar cell patterned with graded photonic super- 4 crystals (Manuscript ID: photonics-248146)

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15 **Abstract:** For the first time, we have studied the light trapping enhancement in newly discovered
16 graded photonic super-crystals with dual periodicity and dual basis. Broadband, wide incident
17 angle, and polarization independent light trapping enhancement was achieved in silicon solar cells
18 patterned with the graded photonic super-crystals. **The design of the graded photonic super-crystals**
19 **is flexible and efficient as they can be realized by multi-beam interference.** The optical response of
20 the patterned silicon solar cell retains the Bloch-mode resonance; however, the light absorption is
21 greatly enhanced in broadband wavelengths due to the graded, complex unit super-cell
22 nanostructures, leading to the overlap of Bloch-mode resonances. The broadband, wide angle light
23 coupling and trapping enhancement mechanism are understood to be due to the spatial variance of
24 the index of refraction due to varying filling fraction, dual-basis, and varying lattice constants in
25 different directions.

26 **Keywords:** light trapping; photonic crystals; micro- and nano-structured materials; photovoltaic
27 devices

28

29 1. Introduction

30 When solar cells become thinner, ultrathin materials used in the solar cells become almost
31 transparent and have low efficiencies to trap or absorb light. Advanced light trapping techniques are
32 required for solar cells to achieve high efficiencies. Nanostructures have been used to improve light
33 coupling from free space to silicon based photovoltaic solar cell devices and incorporate light-
34 trapping functionality in the solar cell in order to increase the light absorption of the device. Various
35 nanostructures have been proposed and their mechanisms for light absorption enhancement have
36 been investigated, which include surface plasmon induced enhancement [1], nanowire-based light
37 trapping [2-4], back-reflector-based multiple pass absorption [5-9], and one-dimension and two-
38 dimension photonic crystal-based structural resonances [10-14]. Photonic crystals are periodic
39 structures that offer powerful photon control and manipulation capability [10-11]. The structural
40 resonances in simple photonic crystals offer a series of sharp resonances at a specific wavelengths
41 and angles [10-14]. By incorporating dual lattice or so-called superlattice structures into photonic
42 crystals, the spectral response of the photon-lattice interactions can be broadened [15-18].

43 Small amounts of position disorder in photonic lattices can be used to control light coupling into
44 the lattice by controlling the ratio of Bloch-mode resonances to Anderson-localized modes and to
45 improve light trapping in solar cell devices [15,19,20]. Furthermore, designs using randomly or quasi-
46 randomly textured surfaces have achieved both high efficiency light trapping through coherent light
47 scattering and broadband, wide angle optical properties due to the introduced disorders [15,21-24].
48 **The strong Bloch mode-based resonant absorption in narrow bandwidths and broadband absorption**
49 **with enhancement can be balanced by controlling the degree of disorder in the nanostructured**
50 **device; however, this can be difficult [19,21].**

51 In this paper, for the first time, we have studied light trapping enhancement in silicon patterned
52 with nanostructures that can be considered as a hybrid of photonic crystals and disorder. **High**
53 **resonance absorption and broadband optical response were simultaneously achieved in silicon**
54 **patterned with graded photonic super-crystals (GPSCs) with dual basis and dual periodicity.** In
55 contrast to wavelength broadening due to disorder in the nanostructures, the broadband and wide
56 angle optical response in the newly discovered GPSCs was achieved through gradient refractive
57 index in dual basis nanostructures.

58 2. Description of Graded Photonic Super-crystals and Simulation Methods

59 Using the results of Oskooi et al [19] as a comparison, similar solar cell structures were used in
60 the simulations with the GPSC pattern instead of a disordered photonic crystal pattern. As shown in
61 Figure 1(a), a 300 nm indium-tin oxide (ITO) ($n=1.8$) covered glass slide can be spin-coated with a
62 positive resist, then patterned with the newly discovered GPSC [25,26] as shown in Figure 1(b), and
63 finally etched to a depth of 200 nm. A gold film is then deposited and the positive resist is lifted off.
64 The gold film will initiate the silicon growth for 200 nm (fill the groove of the pattern) [27]. Gold is
65 again deposited the whole flat surface to initiate the silicon film growth for a thickness of 300 nm.
66 Then the gold film is etched away. A 50 nm silver film is deposited as the metal back-reflector
67 following 100 nm ITO deposition.

68 The lattice constant, a , was used to describe the feature size of the nanostructure as shown in
69 Figure 1(b) for a $12a \times 12a$ unit super-cell of GPSC. If **the parameter a is less than 500 nm, electron-**
70 **beam lithography can be used for the fabrication GPSC pattern. An SEM of a fabricated GPSC using**
71 **electron-beam lithography in the positive resist PMMA is shown in Figure 1(c). When the lattice**
72 **constant is above 500 nm, eight-beam interference lithography (four inner beams with an interference**
73 **angle of α and four outer beams with an interference angle of β) can be used to fabricate the pattern**
74 **[26,27]. A GPSC with a unit super-cell size of $26a \times 26a$ (with $a=1100$ nm) has been fabricated using**
75 **eight-beam interference [27]. The unit super-cell size can be controlled by the selection of angles α**
76 **and β [27]. It is easier to fabricate a GPSC with a large unit super-cell than one with a small unit super-**
77 **cell. However, it is difficult to have a high resolution simulation for a GPSC with a large unit super-**
78 **cell. We expect that a GPSC with a unit super-cell of $12a \times 12a$ can be achieved with experimental**
79 **fabrication and high resolution simulation.** The authors acknowledge that the lattice constant, a , is
80 not in the right range; however, this paper focuses on the simulations only and the experimental
81 results will be published in the future when the GPSC with the desired structural parameters has
82 been fabricated and characterized.

83 In the simulation, the eight-beam interference patterns were converted to binary **complex**
84 **dielectric** structures by comparing the interference intensity, $I(r)$, with a threshold intensity, I_{th} (as a
85 percentage of maximum intensity), using the following step functions: **complex** dielectric
86 material=silicon, when $I < I_{th}$, and **complex** dielectric material=ITO when $I > I_{th}$. **A complex dielectric**
87 **function of amorphous silicon should be used.** However, the **complex dielectric function of**
88 **amorphous silicon depends on the growth conditions [28]** and no tabulated data was found.
89 Furthermore, this paper will compare structural resonance difference between silicon patterned with
90 a GPSC and silicon patterned with a conventional photonic crystal. The selection of optical
91 parameters of silicon will have less effects on the resonance peaks than the structural parameters of
92 the photonic crystals. In this paper, we have used tabulated data of the optical properties (refractive
93 index and extinction coefficient) of crystalline silicon from 250 to 1000 nm [29]. Nano-structuring of

94 silicon using GPSCs and conventional photonic crystals show little difference in improving light
 95 trapping in the high absorption region. Thus the simulations are presented in the wavelength range
 96 between 500 and 1000 nm. Figures 1(b) shows a permittivity structure output that is used to check
 97 the accuracy of the simulation program in MIT MEEP.

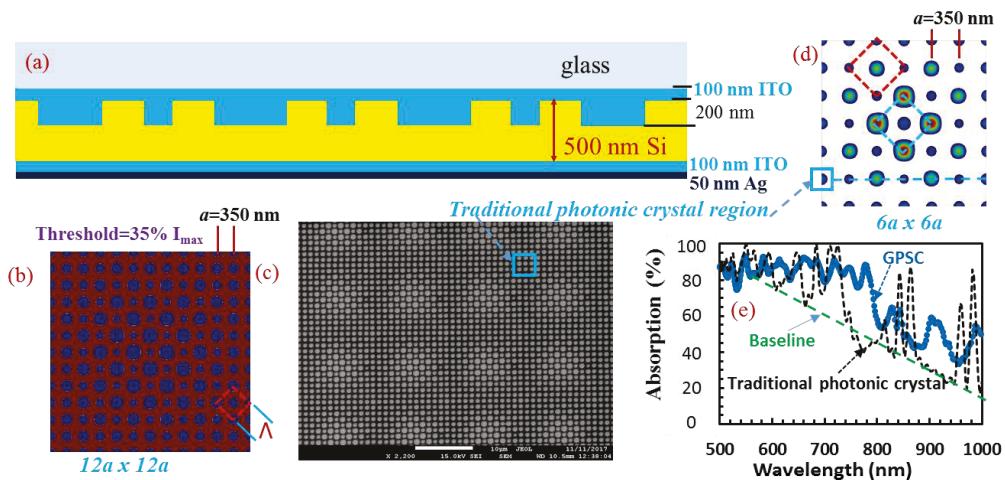


Figure 1. (a) Schematic of a silicon photovoltaic device patterned with a GPSC on a silicon (Si) absorbing layer. (b) Permittivity structures output from MIT's MEEP simulation with a unit super-cell size of $12a \times 12a$ ($a=350$ nm, the red region is silicon and the blue region is ITO). (c) SEM of fabricated GPSC in PMMA using e-beam lithography. (d) A unit super-cell size of $6a \times 6a$ ($a=350$ nm) formed by eight-beam interference with a threshold intensity $I_{th}=30\%$ of the maximum. (e) An eight-beam interference pattern with a unit super-cell of $6a \times 6a$ ($a=350$ nm). The dashed squares link lattices at corners that belong to one set of graded lattice. (e) Absorption spectra for silicon patterned with GPSC with a unit cell of $6a \times 6a$, and with a traditional photonic crystal with a unit cell size of 350×350 nm 2 with silicon and a circle of ITO with 145 nm diameter in the center. The dashed green line indicates the baseline of absorption for eye guidance.

98 The simulations of absorption fraction in silicon patterned with the GPSC and conventional
 99 photonic crystal were performed using MIT's open-source finite-difference time-domain (FDTD)
 100 software tool [30] via the Simpetus Electromagnetic Simulation Platform in Amazon Web Services
 101 (AWS). Due to the large unit super-cell size, 36-core virtual machines in AWS were selected for
 102 parallel computations. For the unit super-cell size of $12a \times 12a$, a resolution as large as 22 can be used
 103 corresponding to an estimated mesh size of $350/22=15.9$ nm (about 1/31 of 500 nm light wavelength).
 104 For an accurate comparison of the silicon patterned with the GPSC and conventional photonic crystal,
 105 a unit super-cell size of $6a \times 6a$ as shown in Figure 1(d) and a resolution as large as 40 is used
 106 corresponding to an estimated mesh size of $350/40=8.8$ nm. For the structure in Figure 1(a) with a
 107 silver layer as a reflector, the transmission $T(\lambda)$ is zero. In the simulation, only the reflection $R(\lambda)$ was
 108 calculated and plotted. Using $A(\lambda)=1-T(\lambda)-R(\lambda)$, an absorption was obtained. An integrated
 109 absorption can be calculated using the tabulated AM 1.5G solar spectrum. However, this was not
 110 done in order to compare results with others under the same conditions [19-20]. Sunlight is almost
 111 unpolarized. To best explain the simulations, the average values of the simulations with incident light
 112 polarized in the [0,1] and [1,1] directions can be displayed, but in this study we chose to display them
 113 separately to analyze the polarization effects, which is one of distinguished features of silicon
 114 patterned with GPSC.

115 3. Results

116 Figure 1(d) shows a GPSC with unit super-cell of $6a \times 6a$ ($a=350$ nm) formed by eight-beam
 117 interference. The highest absorption occurs for the interference pattern with threshold intensity at
 118 30% of maximum intensity. Figure 1(e) shows the absorption of 500 nm silicon patterned with a GPSC
 119 with the unit super-cell of $6a \times 6a$ and with $I_{th}=30\%$ of the maximum intensity. The absorption from

120 500 nm silicon patterned with a conventional photonic crystal is also shown in Figure 1(d) for
 121 comparison. The conventional photonic crystal is a square lattice with a uniform pattern (without
 122 gradient structure) as indicated by solid blue square in Figure 1(c). We picked up a pattern (as
 123 indicated by a solid blue square) near the edge of the GPSC and measured the diameter of the pattern
 124 in Figure 1(d). We used a unit cell size of $350 \times 350 \text{ nm}^2$ square of silicon with a circle of ITO of 145 nm
 125 diameter in the center for the conventional photonic crystal. Comparing both spectra, (a) the
 126 absorption in silicon patterned with the conventional photonic crystal shows relatively sharp Bloch-
 127 mode resonance peaks while the absorption spectrum from the silicon with the GPSC shows
 128 smoothed Bloch-mode resonance peaks, similar to the spectra of silicon patterned in photonic crystal
 129 with 3.6% positional disorders in Figure 2 of reference 19; (b) despite the overall decreasing
 130 absorption baseline with increasing wavelength as indicated by the dashed green line in Figure 1(e)
 131 and that in reference 19 by Oskooi et al., the absorption in Figure 1(e) for silicon patterned with the
 132 GPSC is kept at a high level between 500 and 780 nm (i.e. broadband). The broadband light coupling
 133 can be understood by Eq. (1) [31]:

$$134 \quad \frac{2\pi}{\lambda} n_{eff} - \frac{2\pi}{\lambda} \sin(\theta) = G \quad (1)$$

135 where n_{eff} is the effective refractive index of the GPSC, λ is the wavelength in free space, and G is the
 136 reciprocal lattice vector. G can be $2\pi/a$, $2\pi/\Lambda$, $2\pi/(na)$ as defined in Figure 1(b), or others. $\Lambda = \sqrt{2}a$ is
 137 the periodicity in the 45 degree direction relative to the x or y axis and na (n is an integer number) is
 138 the length of the large unit super-cell, e.g. $6a$.

139 The effective refractive index is defined as follows:

$$140 \quad n_{eff} = f \times n_{ITO} + (1 - f) \times n_{silicon} \quad (2)$$

141 Where n_{ITO} is the refractive index of ITO, $n_{silicon}$ is the refractive index of silicon, and f is the fill fraction
 142 of ITO in the GPSC.

143 The GPSC consists of two sets of graded lattices. The dashed squares in Figure 1(d) link the
 144 corner lattices that belong to one set of the graded lattices. For example, the lattice dots linked by the
 145 dashed blue square in Figure 1(d) have a large circle in the center and gradually decrease their sizes
 146 in toward the edge the unit super-cell. The two sets of graded lattices have the same periodicity thus
 147 G parameter in Eq. (1) is the same. However, the fill fraction f is spatially varying. For example
 148 $f=13.5\%$ for a circle of ITO with 145 nm diameter in a $350 \times 350 \text{ nm}^2$ square of silicon at the left side of
 149 the dashed blue line, while $f=8.9\%$ for a $350 \times 350 \text{ nm}^2$ square region in the middle of the line. Due to
 150 the availability of different fill fractions, light wavelengths in a broad range can meet the condition
 151 in Eq. (1). Thus, broadband light trapping enhancement is observed in the simulation for silicon with
 152 the GPSC due to the overlap of Bloch-mode resonances.

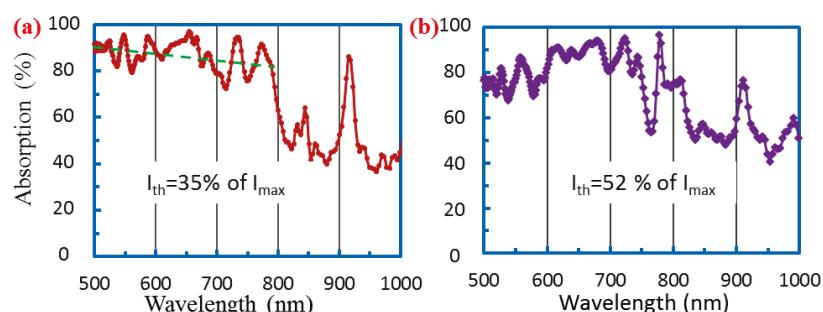


Figure 2. (a) Absorption versus wavelength profile at normal incidence for the silicon patterned with two GPSCs with unit super-cell size of $12a \times 12a$: a GPSC of threshold intensity $I_{th}=35\%$ (a) and a GPSC of threshold intensity $I_{th}=52\%$ (b).

153 The degree of variation in the graded lattices depends on the location of the interference pattern
 154 as described later in the discussion section, and also on the size of unit super-cell. For GPSCs with a
 155 unit super-cell size of $12a \times 12a$, the corner areas in Figure 1(b) and regions indicated by the solid blue

square in Figure 1(c) have a uniform instead of graded lattice and become a conventional photonic crystal. The effect of the conventional photonic crystal can be seen in the absorption spectrum. For silicon patterned with a GPSC with unit super-cell of $12a \times 12a$, a maximum average absorption occurs with $I_{th}=35\%$ of the maximum intensity, as shown in Figure 2(a). Broadband absorption and smoothed Bloch-mode resonance peaks still appear due to the graded regions of the pattern; however, the overall absorption decreases with increasing wavelength, as indicated by the dashed green line, due to the conventional photonic crystal regions. When the threshold intensity is increased from 35% in Figure 2(a) to 52% in Figure 2(b), fill fraction f is decreased and n_{eff} is increased. Therefore, the wavelength for the overall absorption maximum is also red-shifted, from Eq. (1) and (2) and as shown in the figures.

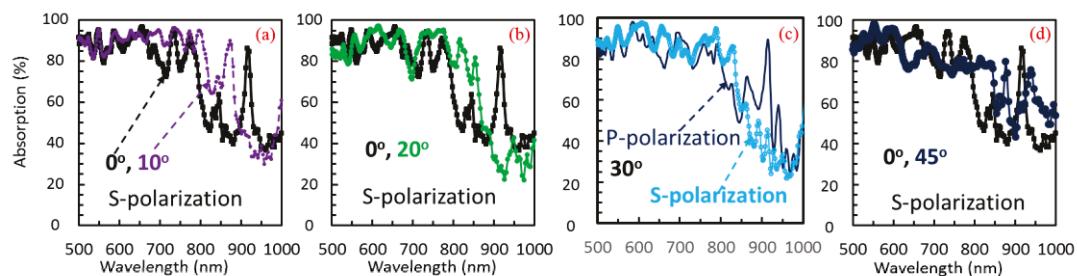


Figure 3. (a-d) Absorption spectra versus wavelength for 500 nm silicon patterned with GPSC with a unit super-cell of $12a \times 12a$ ($a=350$ nm) and with a threshold intensity of 35% of maximum with [1,0] polarized light at normal incidence, compared with the absorption spectrum at four off-normal angles of incidence (zenith angle=10, 20, 30 and 45 degrees respectively) with s-polarization. The absorption from the silicon patterned with the GPSC at an incident angle of 30 degrees with p-polarization is also shown in (c).

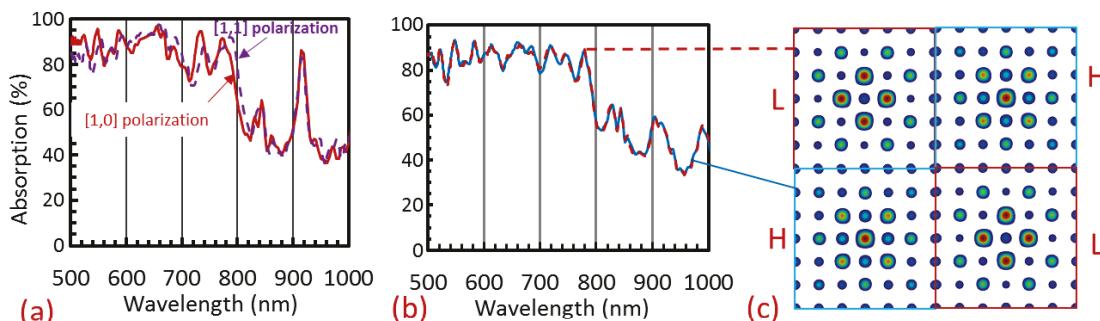


Figure 4. (a) Absorption spectra of light with polarizations in the [1,0] and [1,1] direction incident on silicon patterned with a GPSC with a unit super-cell of $12a \times 12a$ ($a=350$ nm), formed with a threshold of 35% of maximum interference intensity. (b) Absorption spectra of light with polarizations in the [1,0] direction incident on silicon patterned with a GPSC with a unit super-cell of $6a \times 6a$ ($a=350$ nm), formed with a threshold of 30% of maximum intensity for the pattern from the L and H regions in (c). (c) Eight-beam interference pattern with unit super-cell size $6a \times 6a$, showing two distinct regions marked with an L (L for low intensity spot in the center) and an H (H for high intensity spot in the center).

Figure 3 shows the dependence on incident angle for 500 nm silicon patterned with a GPSC with a unit super-cell size of $12a \times 12a$ ($a=350$ nm) and a threshold intensity of 35% of the maximum intensity, for s-polarized light, or [1,0] polarization, with zenith angle of 10, 20, 30 and 45 degrees for Figure 3(a,b,c,d) respectively, and azimuth angle of 90 degrees relative to x-axis for all. Compared with the absorption spectrum at normal incidence ($\theta=0^\circ$), the spectrum with the incident angle of $\theta=10^\circ$ shows higher absorption around 700 nm, high broadband absorption region expanded to 800 nm, and blue-shift of the Bloch-mode resonance peak around 900 nm. The absorption spectra at 20 and 30 degrees show almost the same light absorption as the one at 0 degrees except a broader high absorption region expanded to 800 nm. At an incident angle of 45 degrees, the simulated absorption

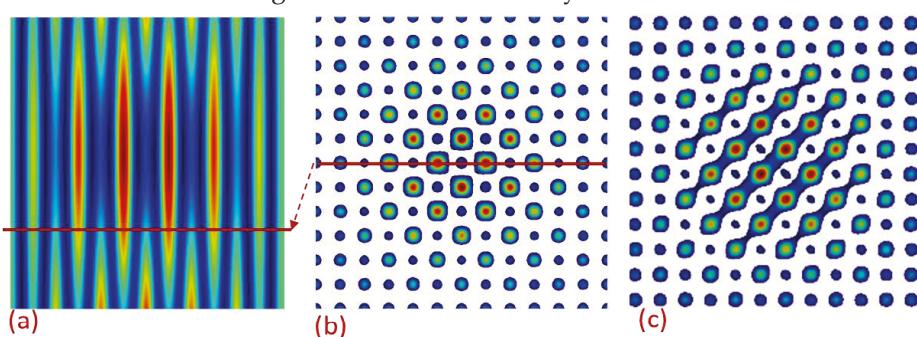
175 decreases above 600 nm. Thus, under the wide angle range of 30 degrees, the absorption is almost
 176 same. For p-polarization at a zenith angle of 30 degrees in Figure 3(c), the absorption peak around
 177 900 nm still appears and the high absorption band is a little bit narrower than that with s-polarization.

178 Figure 4 shows the absorption of light with polarizations in the [1,0] and [1,1] directions incident
 179 on silicon patterned with the GPSC with a unit super-cell size of $12a \times 12a$, formed with the same
 180 threshold of 35% of maximum intensity. We can see from Figure 4 that the absorption spectra are the
 181 same when the light is incident with different polarizations. In the [1,1] direction of the GPSC, the
 182 lattice periodicity is larger, compared with that of the [1,0] direction. The gradient filling fraction in
 183 the [1,1] direction helps couple the light into the patterned structures, with similarly high
 184 enhancement as with [1,0] polarized light, by meeting the conditions in Eq. (1) and (2). Thus, we have
 185 achieved polarization-independent broadband absorption when the silicon solar cell is patterned
 186 with the GPSC.

187 If the GPSC is formed by eight-beam interference as shown in Figure 4(c), there are two “L”
 188 square regions where the central spot has a “low” intensity and two “H” regions where the central
 189 spot has a “high” intensity. If the unit super-cell is large, the “high” or “low” spot in the center is not
 190 critical as there are many lattice spots. However, the “high” or “low” spot in the center might become
 191 crucial in the absorption when the number of lattice spots become small. We simulated the absorption
 192 spectra for silicon patterned with GPSC with a unit super-cell size of $6a \times 6a$ with formed with a
 193 threshold of 30% of maximum intensity, as shown in Figure 4(b). The absorption from “H” region
 194 (dashed red line) is almost identical to that from “L” region (blue line).

195 4. Discussion

196 For photovoltaic devices, nanostructuring the active layers has generated high local density of
 197 optical states in the absorber layer, allowing for more energy to be coupled into the solar cell and
 198 absorbed [10–14]. The appearance of many photonic bands, up to 144, below the photonic band gap
 199 or cavity mode [25] indicates a large density of optical states in the GPSC. When light is coupled into
 200 the structure, we expect resonance modes in the GPSC, waveguide modes in the horizontal plane in
 201 the silicon and ITO regions without GPSC, and leaky resonance modes in the vertical direction. In
 202 the simulations, light source detectors were placed everywhere in a region above the top-layer ITO.
 203 These detectors can collect signals in a large angle of up to 90 degrees, but it cannot collect the
 204 waveguide modes mentioned above. In our simulations of the light extraction in organic light
 205 emitting diode devices [32], the fraction of light power in the waveguide modes is also zero. We have
 206 assumed zero reflection from waveguide modes in this study.



207 **Figure 5.** (a) Cross-section in the x-z plane of the eight-beam interference pattern. (b) Cross-
 208 section in x-y plane of the eight-beam interference patterned as viewed at the location indicated
 209 by the red line in (a). The pattern becomes the one in (c) when the phases of four outer
 210 interference beams are set to be $(0, 0.15\pi, 0, 0.15\pi)$.

211 High resonance absorption and broadband optical response can be further studied via the
 212 optimization of iso-intensity surfaces of the interference pattern, lattice constant, and unit super-cell
 213 size. Figure 5(a) shows a cross-section in x-z plane of the interference pattern. When the sample is
 214 exposed at the location as indicated by the red line, the pattern will look like the one in Figure 5(b)
 215 where the one set of lattice spots is smaller than the other. If the red line is shifted to the bottom of
 216 Figure 5(a), the formed pattern will have a similar spot size in both sets of lattices. If the phases of the

213 four outer interference beams were set to be $(0, 0.15\pi, 0, 0.15\pi)$, the pattern in Figure 5(b) will become
214 the one in (c).

215 **5. Conclusions**

216 For the first time, we have simulated the fraction of absorption of light in silicon solar cells where
217 the silicon is patterned with GPSCs, which are a hybrid of photonic crystals and disorder. Due to the
218 spatially gradient fill fraction of dielectrics and large number of available lattice constants in different
219 directions, we have achieved broadband, wide angle, and polarization-independent absorption
220 spectra. Broadband light trapping enhancement has been explained in terms of overlapping Bloch-
221 mode resonances.

222

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226 performed the simulations; Y.L., S.H. and D.L. analyzed the data; D.L. contributed parallel computation tools;
227 Y.L. and S.H. wrote the paper. All authors read and commented on the manuscript.

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