

Broadband Radiative Heat Transfer Suppression via Dispersion Engineered Metasurfaces

Corresponding Author: Professor Andrea Alu

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript highlights the application of aperiodic distributed Bragg reflectors to realize multiple spectral bandgaps between two metasurface structures to mitigate radiative thermal transport. The design of the structures was enabled by the use of an inverse design protocol using stochastic gradient descent, with the authors demonstrating both a 3-layer pair and a 7-layer pair, demonstrating the higher spectral control obtained via the latter. Overall the results are interesting and the application is in need of such a development, and as such, I believe this work should be published in Nature Communications. However, there are a few things I think the authors should address before publication.

1. This work is similar in approach to reference 53, which used a similar inverse design protocol to engineer Tamm-like thermal emitters, which was led by the second author of this work. As such, it should be highlighted what developments in the inverse design code, methodology and the resulting outputs were made beyond that initial paper. I do not believe this undercuts the novelty, even if they used the exact same code, but if this is a modification or is just a new use of the same code, this should be clearly stated.
2. I think the work could be significantly assisted by having a schematic design figure up front that clarifies the implementation of this approach within the context of this work. e.g., how would such structures help the state of the art? This is somewhat demonstrated in Fig. 1a, but even if they use that, it is introduced really late in the paper and I don't think the intro makes this clear enough as to why this is helpful or impactful.
3. Why are the 7-layer p-polarized results in the SI and not the main text? p-pol would be where we would anticipate any polaritonic behaviors, which considering the SiO₂ substrate has a nice clean reststrahlen band, this could be non-negligible response, yet it is not even discussed in the main text. I strongly believe either the figures should be included, or the reason why they are not included in the main text be stated. Along these same lines, is the substrate quartz or fused silica? With that clean of a reststrahlen band it would appear that there could be polaritonic modes supported, this should be discussed. Within the SiO₂ layers, I would presume this not to be the case considering the amorphous nature, but this is unclear from the discussion. I would ask that the absence of these modes from the response be clarified.
4. In Fig. 3, the authors use different shades of the same color to show the calculated and experimental stacks for both the radiator and absorber structures. I understand the goal, but especially for the 7-layer structure, this is near impossible to evaluate and compare without an exceptional amount of focus and time. I would encourage them to maybe make the radiator curves red and the absorber layers blue to clearly differentiate? It would be good to discuss some of the discrepancies between the designed and fabricated spectra.
5. I would STRONGLY encourage the authors to change the acronym for electrical substitution radiometer (ESR). ESR is broadly used for electron spin resonance and to avoid confusion, this acronym should be changed.

Reviewer #2

(Remarks to the Author)

This manuscript by Jing et al. presents a highly innovative and experimentally rigorous approach to suppressing broadband radiative heat transfer without the use of metallic reflectors. The authors cleverly circumvent the fundamental Bode-Fano and

Rozanov limits by shifting the design paradigm from a single-surface broadband suppression to a dual-surface "spectral misalignment" strategy. By utilizing stochastic gradient descent (SGD) to co-optimize aperiodic a-Si/SiO₂ distributed Bragg reflectors (DBRs), they successfully engineered complementary narrowband resonances that remain thermally decoupled over a wide spectral range. The authors custom-built an electrical substitution radiometer (ESR) to directly quantify the net radiative power between two macroscopic wafers under high vacuum. This direct measurement of a 62.5% reduction in radiative heat exchange compared to fused silica.

Overall, the concept is elegant, the engineering implementation is robust, and the experimental physics are exceptionally solid. There are several critical discrepancies between the theoretical design and the experimental reality that must be addressed.

1. There is a significant gap between the predicted heat transfer suppression (~82%) and the directly measured value (62.5%). In the Supplementary Materials, the authors attribute this ~20% performance drop to the unoptimized radiative contributions in the far-IR range and angular dependencies at higher incidence angles. What if these factors are incorporated into the objective function for a re-optimization? The authors may either (a) provide a revised SGD optimization that integrates the 30-8000 cm⁻¹ spectrum and hemispherical angular integration to show if this is doable theoretically, or (b) provide a rigorous physical justification (e.g., computational cost, material constraints) for why the current design space is restricted.
2. The initial design assumed normal-incidence/diffuse emission to simplify the calculations. However, as shown in Fig. S10, the spectral misalignment degrades significantly at large incidence angles due to phase shifts in the DBR layers. The authors may want to quantify the "angular bandwidth" within which the misalignment remains effective and discuss whether incorporating angular averaging into the SGD cost function would fundamentally alter the thickness distributions.
3. Fig. 4(d) shows that mutual emissivity drops exponentially with the number of layers, yet the authors stopped at 7 layers. The authors mention "fabrication complexity and thermal stress" as limiting factors. To strengthen the manuscript's engineering relevance, the authors should briefly elaborate on this trade-off. What is the critical thickness or layer count where a-Si/SiO₂ films will undergo mechanical failure (delamination/cracking) during thermal cycling? Does the 7-L pair represent the absolute mechanical limit, or just a convenient proof-of-concept?
4. The authors used Stochastic Gradient Descent (SGD). Since the parameter space (layer thicknesses) is highly non-convex with many local minima, how did the authors ensure that SGD converges to a globally optimal (or near-optimal) solution? A brief discussion comparing SGD to global optimizers (like Genetic Algorithms or Particle Swarm Optimization) in the context of this specific multi-layer problem would be beneficial.
5. The authors cite the need for non-metallic radiative suppression in electronic thermal protection and thermoelectric systems. It would be interesting to briefly discuss how this spectral misalignment strategy used in near-field thermophotovoltaics for waste heat recovery (see, for example, APPLIED PHYSICS LETTERS 107, 091106 (2015)), as both aim to suppress sub-bandgap parasitic heat transfer. Would the same design rationale presented here be applicable to the near-field heat transfer as well? What would be different?

Reviewer #3

(Remarks to the Author)

In the present work the authors propose a broadband radiative heat transfer suppression scheme based on two dielectric metasurfaces made of distributed Bragg reflectors. The layer thicknesses of the two stacks are optimized using gradient descent with the goal of reducing radiative exchange between them. Essentially, what the authors are aiming to achieve is to instead of trying to make each surface weakly emitting across the entire infrared band, they design two surfaces that are spectrally incompatible with each other, so that frequencies emitted efficiently by one terminal are, as much as possible, not absorbed efficiently by the other. This reduces the spectral overlap that normally enables radiative heat transfer. This is then verified experimentally.

The overall idea is interesting and potentially useful, especially for applications where metallic coatings are not desirable but suppression of radiative heat transfer is still needed. The use of metasurface based multilayer coatings and the optimization of the two interacting terminals rather than a single surface also represent an interesting design strategy in my opinion. The work therefore points toward an interesting and novel direction for controlling radiative exchange using engineered photonic structures.

However there are several points that should be clarified or strengthened.

My main remark concerns how the paper frames the work relative to bandwidth limits and the discussion of Bode Fano and Rozanov bounds. The manuscript refers to the idea of "going beyond bandwidth constraints" and mentions "circumventing the Bode Fano bound because it applies to continuous bandwidths". However the device considered here is still linear time invariant and passive. As such its scattering response must still obey the usual constraints. Although I am aware that the authors do not explicitly claim to "break these bounds", the current wording may easily lead readers to interpret the results in that way. In addition the manuscript does not provide a quantitative assessment connecting the device performance to those bounds. The discussion therefore remains largely qualitative and speculative. It would be helpful if the authors clarified more precisely what they mean by circumventing bandwidth limits in this context. For instance, the thermal spectrum being integrated in the heat transfer calculation is still "continuous", so the statement that the approach avoids "continuous bandwidth" constraints is not immediately clear.

A second important point is about the consistency between the modeling and the experimentally measured heat transfer performance. The manuscript states suppression levels inferred from emissivity spectra as well as suppression levels

obtained from direct heat transfer measurements. However the two estimates differ noticeably, especially for the stronger performing multilayer design. The authors suggest possible explanations such as spectral contributions outside the original design window, but at present the central performance metric is not predicted consistently by the modeling framework used during the inverse design stage. To me, this leaves the impression that the optimization target is a "proxy" for the actual measured quantity rather than a direct prediction of it. I think strengthening the link between the design objective and the experimentally measured heat transfer would make the central claim more convincing.

Related to this, part of the modeling framework relies on treating the structures as effectively opaque when extracting emissivity from reflection measurements. However the manuscript also notes that this assumption breaks down in certain spectral regions where transmission becomes non negligible. When the structures are not strictly opaque, the relationship between reflection and emissivity becomes more complicated and this can affect both the emissivity spectra used for the analysis and the calculation of mutual emissivity between the two terminals. Since the suppression mechanism relies on carefully shaping the spectral response, such modeling assumptions could influence the predicted heat transfer suppression. I suggest that the authors should address this point more carefully in the modeling and design procedure would improve the internal consistency of the analysis.

This is somewhat more speculative, but it may be useful to consider possible alternative approaches that could achieve a similar effect with simpler design procedures. For instance, one could also use two chirped reflectors designed to provide broadband reflection, but with slightly different reflection or emissivity profiles across the spectrum. Here the suppression of radiative exchange would arise from a broad spectral mismatch between the two surfaces, rather than from a "piecewise" mismatch created by multiple narrow resonances. Such a strategy might be significantly easier to design since chirped multilayer reflectors are well understood and can often be obtained using straightforward analytical design rules without the need for optimization. Also, designs that rely less on narrow resonances and resonance alignment may be more robust to fabrication imperfections or layer thickness variations. Perhaps it would be helpful if the authors clarified how the present approach compares with such alternatives. In particular, is the main advantage of the proposed design that it can achieve the desired suppression with fewer layers or reduced total thickness, or that it offers a higher degree of control over the spectral response?

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I have reviewed the updated documents and response letter. I appreciate the significant efforts the author's made in response to all three reviewer's comments and believe the manuscript is ready for publication.

Reviewer #2

(Remarks to the Author)

The authors addressed all comments and I think the work can be published.

Reviewer #3

(Remarks to the Author)

The authors have satisfactorily addressed the reviewer comments and concerns.

In line with my previous report, the main idea is interesting and useful, especially for non-metallic applications where suppression of radiative heat transfer is strongly needed. As such, the work points towards a novel direction for controlling radiative exchange using artificial photonic structures.

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Response to the Reviewers' Comments for Manuscript NCOMMS-26-014939 'Broadband Radiative Heat Transfer Suppression via Dispersion Engineered Metasurfaces'

We thank all the reviewers for their insightful comments and suggestions during the review process. We believe to have addressed all of them in this document and in the revised manuscript as appropriate (highlighted in blue). More details and supporting evidence can be found in the Supplementary Materials.

To address the main comments with greater clarity, we have included the key revision, Fig. 4(c)-(e), here. Specifically, it presents updated data showing improved agreement between direct measurements and indirect estimations. The corresponding detailed analysis is provided in SM Section 7, which includes the systematic discussion on the considered wavelength range, non-opaqueness, and angle-dependent emission. For comments related to these points, we refer the Reviewers to the corresponding content for more details.

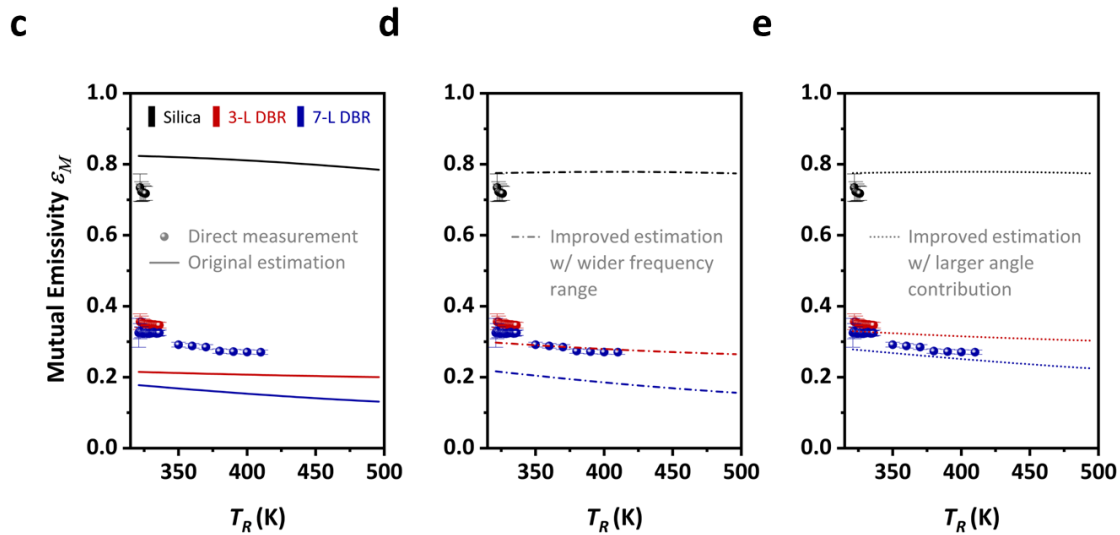


Fig. 4 (c) Mutual emissivity versus radiator temperature for: bare silica substrate (black), 3-L DBR pair (red), and 7-L DBR pair (blue). Scattered dots represent experimental data obtained by electrical substitution radiometer measurement, whereas solid lines denote indirect estimations derived from measured emissivity. (d) Dash-dot lines denote the improved estimation using extended-range absorptivity data. (e) Dotted lines denote the further improved estimation by accounting for the large-angle contribution.

Reviewer #1 (Remarks to the Author):

This manuscript highlights the application of aperiodic distributed Bragg reflectors to realize multiple spectral bandgaps between two metasurface structures to mitigate radiative thermal

transport. The design of the structures was enabled by the use of an inverse design protocol using stochastic gradient descent, with the authors demonstrating both a 3-layer pair and a 7-layer pair, demonstrating the higher spectral control obtained via the latter. Overall the results are interesting and the application is in need of such a development, and as such, I believe this work should be published in Nature Communications. However, there are a few things I think the authors should address before publication.

- 1. This work is similar in approach to reference 53, which used a similar inverse design protocol to engineer Tamm-like thermal emitters, which was led by the second author of this work. As such, it should be highlighted what developments in the inverse design code, methodology and the resulting outputs were made beyond that initial paper. I do not believe this undercuts the novelty, even if they used the exact same code, but if this is a modification or is just a new use of the same code, this should be clearly stated.*

Response: We appreciate the reviewer's positive comments, the recommendation for publication, and the constructive suggestions.

We have stated more clearly in the manuscript what has been modified from the original inverse design flow.

"In contrast to our previous research using the same methodology, where the emissivity of a single emitter was optimized, the present development moves toward end-to-end optimization. By assigning appropriate temperature-dependent Planck distribution weights to each frequency, the objective function becomes the radiative exchange power between the emitter and absorber, which is directly calculated and optimized."

- 2. I think the work could be significantly assisted by having a schematic design figure up front that clarifies the implementation of this approach within the context of this work. e.g., how would such structures help the state of the art? This is somewhat demonstrated in Fig. 1a, but even if they use that, it is introduced really late in the paper and I don't think the intro makes this clear enough as to why this is helpful or impactful.*

Response: We thank the reviewer for the suggestion. We have moved the revised conceptual schematic to the front to better highlight the novelty of this work, specifically its realization of broadband radiative transfer suppression optimization using compact all-dielectric metasurfaces—unlike traditional thermophotonic platforms, which are limited to single-stopband or narrowband behavior when benchmarked against broadband radiative exchange between SiO₂ surfaces.

3. *Why are the 7-layer p-polarized results in the SI and not the main text? p-pol would be where we would anticipate any polaritonic behaviors, which considering the SiO₂ substrate has a nice clean reststrahlen band, this could be non-negligible response, yet it is not even discussed in the main text. I strongly believe either the figures should be included, or the reason why they are not included in the main text be stated. Along these same lines, is the substrate quartz or fused silica? With that clean of a reststrahlen band it would appear that there could be polaritonic modes supported, this should be discussed. Within the SiO₂ layers, I would presume this not to be the case considering the amorphous nature, but this is unclear from the discussion. I would ask that the absence of these modes from the response be clarified.*

Response: We appreciate the reviewer's comments and suggestions. We have clarified in the manuscript that the substrate used here is fused silica. We agree that p-polarized results are critically associated with the polaritonic behaviors of SiO₂. Due to space limitations, we have revised Fig. 2 to present polarization-averaged dispersion results for the 7-layer pair, primarily to highlight misaligned resonances rather than showing both polarization-resolved datasets.

In the manuscript, we have added: "Given the limited angular dispersion within the target frequency/angle design space, the normal-incidence emissivity (approximated as diffuse emission) provides a good estimate of the net radiative heat transfer for our demonstration. However, near the SiO₂ Reststrahlen band ($\sim 1100\text{ cm}^{-1}$), even with polarization averaging, Fig. 2(d)–(g) reveal clear dispersive behavior with enhanced emission at large angles. This behavior stems from the phonon-polaritonic response of SiO₂, where LO-TO (longitudinal optical–transverse optical) phonon splitting results in negative permittivity within this spectral region^[59, 62]. These modes respond exclusively to p-polarized (TM) fields, as confirmed by polarization-resolved measurements (Supplementary Materials, Section 3). In the Si/SiO₂ multilayer stack, they couple to radiative channels and become increasingly prominent at large emission angles due to improved momentum matching. Consequently, dispersion becomes non-negligible at small separations, where large-angle contributions play a significant role in heat transfer. Mitigating this effect is therefore necessary and is addressed in the next section."

We also added an entire Section 3 in the SM for all the polarization-resolved data along with the discussion on the polarization-dependent effects. We refer the reviewer to the corresponding section for details.

4. *In Fig. 3, the authors use different shades of the same color to show the calculated and experimental stacks for both the radiator and absorber structures. I understand the goal, but especially for the 7-layer structure, this is near impossible to evaluate and compare*

without an exceptional amount of focus and time. I would encourage them to maybe make the radiator curves red and the absorber layers blue to clearly differentiate? It would be good to discuss some of the discrepancies between the designed and fabricated spectra.

Response: We thank the reviewer for the suggestion. Since direct comparisons among the benchmark, 3-L pair, and 7-L pair are presented in Fig. 3(d) and Fig. 4(c)-(e), we retained the same color palette to ensure consistent color labeling throughout the manuscript. However, we have recalculated all data using more accurate structural parameters (including the presence of the substrate) and material properties, which has improved both the agreement between experiment and calculation and the overall visual clarity. We have also added the following discussion to the manuscript: “The remaining discrepancy between calculation and experiment is attributed to experimental factors, such as temperature mismatch, atmospheric absorption, and partial transparency in certain spectral windows.”

5. *I would STRONGLY encourage the authors to change the acronym for electrical substitution radiometer (ESR). ESR is broadly used for electron spin resonance and to avoid confusion, this acronym should be changed.*

Response: We thank the reviewer for the suggestion. To avoid the confusion, we removed the acronym for electrical substitution radiometer in the manuscript.

Reviewer #2 (Remarks to the Author):

This manuscript by Jing et al. presents a highly innovative and experimentally rigorous approach to suppressing broadband radiative heat transfer without the use of metallic reflectors. The authors cleverly circumvent the fundamental Bode-Fano and Rozanov limits by shifting the design paradigm from a single-surface broadband suppression to a dual-surface "spectral misalignment" strategy. By utilizing stochastic gradient descent (SGD) to co-optimize aperiodic α -Si/SiO₂ distributed Bragg reflectors (DBRs), they successfully engineered complementary narrowband resonances that remain thermally decoupled over a wide spectral range. The authors custom-built an electrical substitution radiometer (ESR) to directly quantify the net radiative power between two macroscopic wafers under high vacuum. This direct measurement of a 62.5% reduction in radiative heat exchange compared to fused silica.

Overall, the concept is elegant, the engineering implementation is robust, and the experimental physics are exceptionally solid. There are several critical discrepancies between the theoretical design and the experimental reality that must be addressed.

- 1. There is a significant gap between the predicted heat transfer suppression (~82%) and the directly measured value (62.5%). In the Supplementary Materials, the authors attribute this ~20% performance drop to the unoptimized radiative contributions in the far-IR range and angular dependencies at higher incidence angles. What if these factors are incorporated into the objective function for a re-optimization? The authors may either (a) provide a revised SGD optimization that integrates the 30-8000 cm⁻¹ spectrum and hemispherical angular integration to show if this is doable theoretically, or (b) provide a rigorous physical justification (e.g., computational cost, material constraints) for why the current design space is restricted.*

Response: We thank the reviewer for the positive comments and constructive suggestions.

We have provided an updated theoretical design in SM Section 5 that covers different frequency ranges, using material constants from the literature. Importantly, covering long wavelength range in the optimization is challenging **because the material resonances are difficult to characterize, and the material loss (e.g., due to optical phonons) tends to vary with different deposition recipes**. Therefore, lacking the capability to accurately model the materials in the experiment—and consequently the entire multilayer system—we did not pursue experimental efforts to cover the wider frequency range. The detailed discussion on the angular dispersion effect is presented in the response to the next comment.

Section 5: Effect of material choices

The performance analysis shows that material resonances of SiO₂ dominating the long-wave infrared range prevent the algorithm from effectively reducing optical crosstalk, even when optimized by considering different frequency ranges, as shown in Fig. S11. Alternatively, the use of lossless materials such as Ge and ZnSe could further improve the suppression performance, as also shown in Fig. S11. However, the point we would like to highlight here is that even with polaritonic materials within the frequency range of interest, we can still find a proof-of-concept optimal suppression given all the constraints.

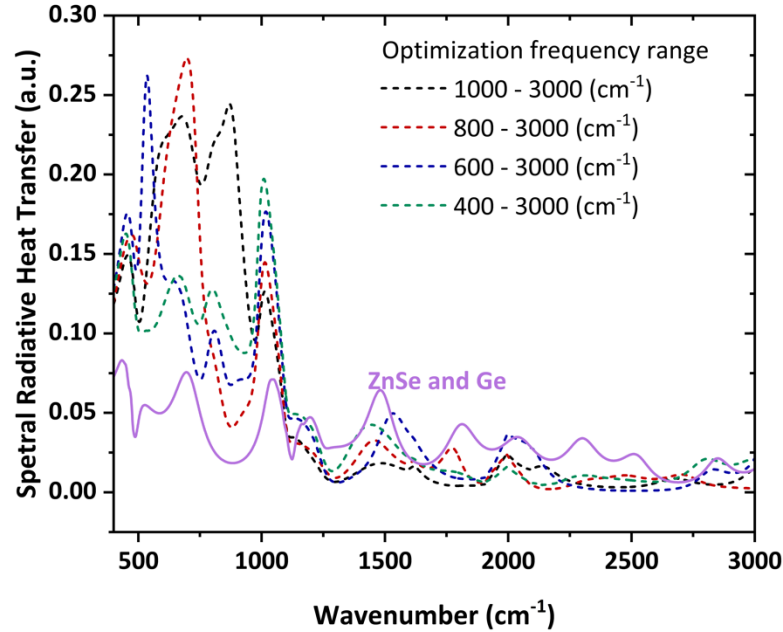


Fig. S11 Spectral radiative heat transfer of 7-L DBR pair optimized using different frequency ranges and constituent materials.

2. The initial design assumed normal-incidence/diffuse emission to simplify the calculations. However, as shown in Fig. S10, the spectral misalignment degrades significantly at large incidence angles due to phase shifts in the DBR layers. The authors may want to quantify the "angular bandwidth" within which the misalignment remains effective and discuss whether incorporating angular averaging into the SGD cost function would fundamentally alter the thickness distributions.

Response: We appreciate the reviewer's insightful suggestion and agree that the angular responses were not sufficiently emphasized in the original submission. To address this, we have expanded the discussion to better highlight the complexity of angular dependence and its impact on net heat transfer. Accordingly, we have evaluated a two-angle averaged effective emissivity to close the gap between the indirect performance prediction and direct

measurement. In addition, we have conducted further analysis showing that p-polarized light contributes significantly to heat transfer at large angles—a fundamental property of this system. Detailed analyses are provided below.

- 1) First of all, we calculated the view factor distribution weights versus angle for two parallel plates according to Eq. S5. The result is presented in Fig. S3 in SM, Section 1. As illustrated, when the gap distance is very small, a sufficiently fine numerical discretization is required to accurately capture the angular distribution of emissivities. Such refinement demands substantial computational resources because the computational cost increases quadratically with the mesh resolution; otherwise, accuracy is severely compromised, leading to numerical fluctuations shown in Fig. S3. Despite these numerical fluctuations, the results clearly show that the contribution from large-angle emission increases as the gap decreases. For small gap sizes, large-angle emission becomes non-negligible and ultimately dominates the response. For instance, when the distance is less than 1 inch, emissivities at angles beyond 75° play a non-negligible role.

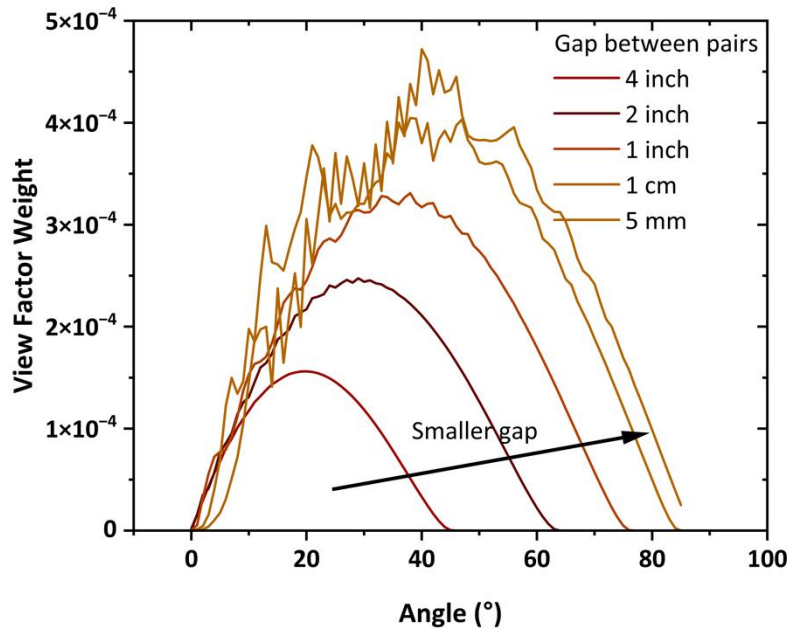


Fig. S3 dA_1 -weighted differential view factor $dF_{1\rightarrow 2}$ distribution.

- 2) Then, in the revised Fig. 4 and SM, Section 7.3, we discuss the effect of large-angle contributions in detail to resolve the discrepancy. In brief, by **incorporating angular averaging** with an assumed "angular bandwidth" of 50° , we have significantly improved the agreement between indirect estimations and direct measurements. As a result, we now have a much clearer picture of the importance of angular dispersion. Future optimizations can be performed by incorporating long-wavelength and angle-dependent emissivities into the modeling for accurate performance prediction and reliable suppression design.

- 3) While the discrepancy between the indirect performance prediction and direct measurement has been resolved, we further investigated whether the suppression performance of this system can be improved. To this end, we performed a series of optimizations targeting reduced optical crosstalk at different angles. However, the crosstalk in p-polarization at high angles could not be effectively suppressed (Fig. R1). This behavior arises from the phonon-polaritonic nature of the Si/SiO₂ structure, as analyzed in detail in Section 3 of the Supplementary Material. Consequently, this **imposes a fundamental limitation on the suppression performance of this structure**, and we made no further experimental efforts to pursue better performance. In our response to Comment 1 and in Section 5 of the SM, we have demonstrated that by selecting appropriate constituent materials, the system can achieve more effective suppression of radiative heat transfer.

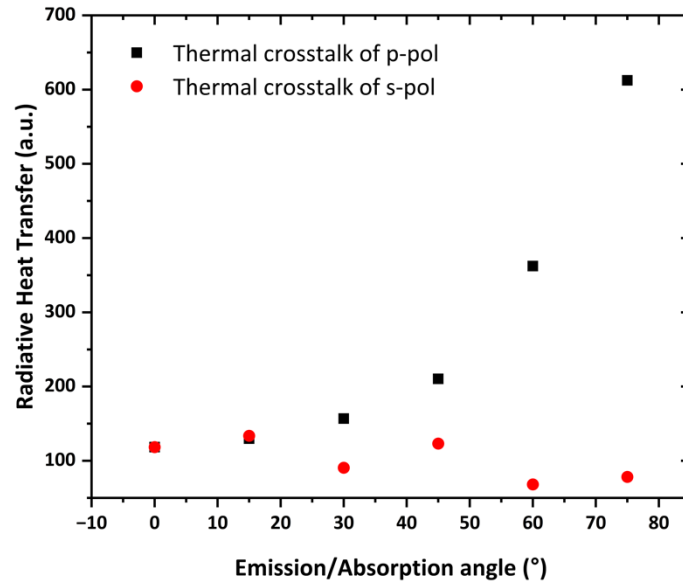


Fig. R1 Polarization-resolved radiative heat transfer vs angle.

SM Section 7.3 is attached here:

Section 7.3 Effect of angle-dependent emission

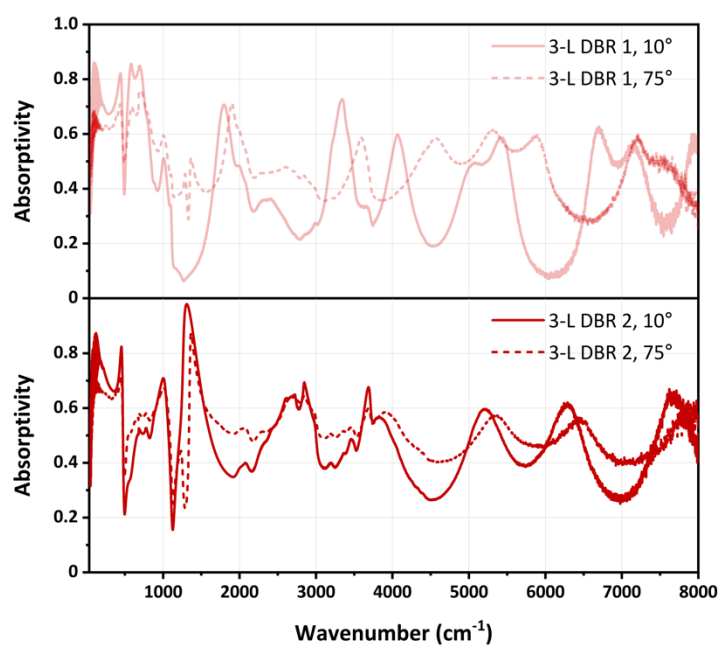
We then revisited the assumption of angle independence (diffuse emissivity) and conducted extended-range FTIR absorptivity measurements at 75° to better understand the remaining discrepancy. Figs. S22–S23 show the measured spectral absorptivity at 10° and 75°. The absorptivity at a higher angle of incidence differs significantly from that at (near) normal-incidence, and the misalignment of emissivity spectra degrades, thereby increasing the overall mutual emissivity. This result is consistent with the direct radiative heat transfer measurements. As shown by the dashed curves in Fig. S24, the mutual emissivity predictions based on the 75° data now overestimate the radiative heat transfer. The reasons,

which have been explained in Section 3, are primarily attributed to the polaritonic behavior of SiO₂ under p-polarization, where a clear shift in dispersion is observed around 50°. To account for this, we calculate two-angle averaged emissivity using two piecewise data points at 10° and 75° with a critical separation angle of 50° as follows. According to Eq. (S4) and Eq. (S5), we obtain:

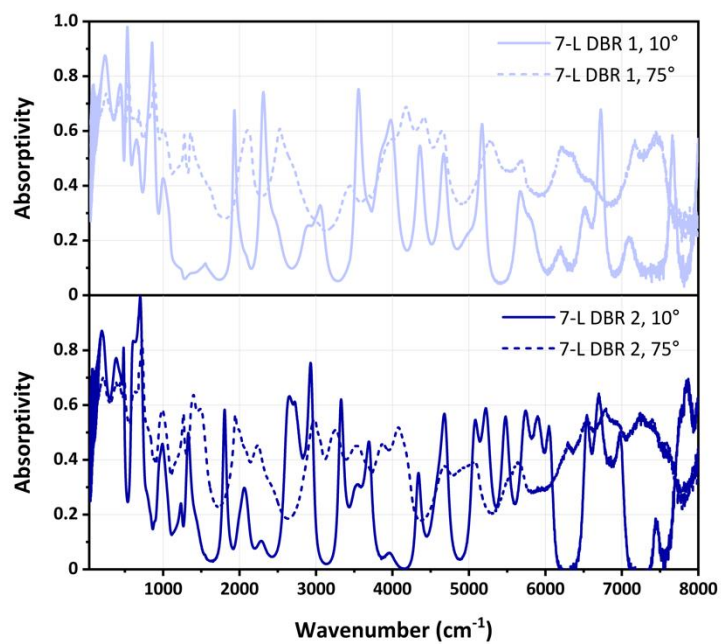
$$\begin{aligned}
 q_{\tilde{\nu}} &= \iint_0^{90^\circ} \varepsilon_M(\tilde{\nu}, \theta) (\Theta(\tilde{\nu}, T_1) - \Theta(\tilde{\nu}, T_2)) \cos\theta \sin\theta d\theta d\phi \\
 &= 2\pi (\Theta(\tilde{\nu}, T_1) - \Theta(\tilde{\nu}, T_2)) \left(\int_0^{50^\circ} \varepsilon_M(\tilde{\nu}, 10^\circ) \cos\theta \sin\theta d\theta + \int_{50^\circ}^{90^\circ} \varepsilon_M(\tilde{\nu}, 75^\circ) \cos\theta \sin\theta d\theta \right)
 \end{aligned}
 \tag{S17}$$

The overall mutual emissivity can then be calculated accordingly. **Fig. S24** demonstrates that this approach significantly improves agreement with the direct measurement, also shown in **Fig. 4(c)** of the main text. Therefore, we conclude that the diffuse approximation used in the optimization procedure becomes less accurate when the material is dispersive, particularly at small gap distances where large-angle contributions are more significant.

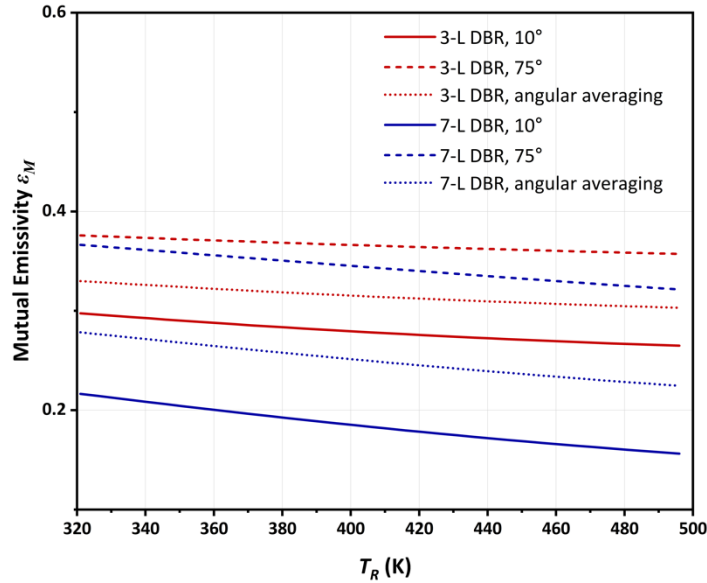
In summary, the above analysis demonstrates that the primary cause of the discrepancy between direct measurement and indirect estimations is the incomplete consideration of the full frequency range and angular dispersion. Consequently, future optimizations can be substantially improved by incorporating these factors and selecting appropriate materials.



[Fig. S22 Absorptivity spectra of 3-L DBR pair at 10° and 75°.](#)



[Fig. S23 Absorptivity spectra of 7-L DBR pair at 10° and 75°.](#)



[Fig. S24 Calculated mutual emissivity of 3-L DBR pair and 7-L DBR pair using different angular contributions.](#)

3. *Fig. 4(d) shows that mutual emissivity drops exponentially with the number of layers, yet the authors stopped at 7 layers. The authors mention "fabrication complexity and thermal stress" as limiting factors. To strengthen the manuscript's engineering relevance, the authors should briefly elaborate on this trade-off. What is the critical thickness or layer count where α -Si/SiO₂ films will undergo mechanical failure (delamination/cracking) during thermal cycling? Does the 7-L pair represent the absolute mechanical limit, or just a convenient proof-of-concept?*

Response: We thank the reviewer for highlighting the important manufacturability trade-off implied by Fig. 4(d), now is Fig. 3(e).

- 1) The 7-L pair is not presented as a fundamental mechanical limit. Rather, it was selected as a **high-yield, proof-of-concept** that (A) is straightforward to fabricate reproducibly with our process flow, (B) remains robust under the operational temperature window explored here (320–500 K), and (C) already achieves large validated suppression. As shown in Fig. 3(e), the 7-L design achieves an 82% reduction in mutual emissivity compared to bare silica, **while adding 10 more layers (i.e., increasing to 17 layers) yields only a modest 6% improvement**. This substantial performance, realized within a simple fabrication framework, presents compelling opportunities for thermal engineering while rendering additional layers unnecessary—although increasing the layer count remains feasible.

- 2) For an a-Si/SiO₂ multilayer stack, the mechanical risk increases with total thickness because residual stress accumulates. Furthermore, during thermal cycling, mismatch in thermal expansion between the multilayer stack and the substrate increases the driving force for cracking and interfacial delamination. Therefore, **the “critical” thickness/layer count is not a universal number**; it depends strongly on (A) deposition method and parameters^{1,2}, (B) intrinsic stress of each film, (C) substrate choice and surface preparation/adhesion (which we observed in our case that fused silica substrate outperforms quartz, and piranha cleaning of the substrate is beneficial to promote the adhesion with deposited films as well), (D) thermal cycling protocol, and (E) whether stress-relief treatment (e.g., anneal) is used. Therefore, **determining the absolute mechanical limit and critical thickness/layer count requires a very delicate systematic investigation**, which is outside the scope of the present proof-of-concept demonstration.

In the manuscript, we have revised as follow: “Fig. 3(e) shows that the mutual emissivity (calculated by normalizing radiative heat transfer to blackbody exchange) exhibits an exponential decay with respect to the number of DBR layers in the design. However, practical implementation requires balancing this improvement against the increased fabrication complexity and thermal stress at higher layer counts. In this work, we selected a 7-L design as a compact, high-yield proof-of-concept that maintains mechanical robustness while already providing strong suppression—adding 10 more layers (i.e., increasing to 17 layers) yields only a modest 6% improvement.”

4. *The authors used Stochastic Gradient Descent (SGD). Since the parameter space (layer thicknesses) is highly non-convex with many local minima, how did the authors ensure that SGD converges to a globally optimal (or near-optimal) solution? A brief discussion comparing SGD to global optimizers (like Genetic Algorithms or Particle Swarm Optimization) in the context of this specific multi-layer problem would be beneficial.*

Response: We thank the reviewer for pointing this out.

We have added the following to the manuscript: “Although all gradient descent-based algorithms are local optimizers, the SGD and additional strategies employed here reduce the possibility of getting trapped in poor local minima within this non-convex parameter space, thus offering a good balance between computational cost and accuracy (a detailed discussion of the optimization algorithm is provided in Supplementary Materials, Section 2).”

We also added an entire Section 2 in SM for detailed discussion, which is shown here as well:

Section 2: Discussion on the optimization algorithms

As the optimization space is highly non-convex, the optimized structures can easily become trapped in poor local minima. However, theoretical studies suggest that with stochastic gradient descent (SGD), the final results tend to settle in a "flat valley" of the parameter space rather than sharp, poor local minima³⁻⁶. To improve the chance of finding near-optimal solution, we run a batch of 10–30 optimizations per task, record the final figure of merit (suppression of heat transfer in this case), and select the best outcome. Because the initialization is random, this multiple-restart strategy helps us avoid poor local minima. Here we provide a histogram (Fig. S4) from 100 optimizations of a representative task—heat transfer suppression using 7-layered DBR pairs. Statistically, we find that approximately 20% of the time, SGD returns "good" local minima, defined as those within 10% of the best result. For example, in this case, if the best result falls within the range of 65–70, then radiative transfer values of 70–75 are considered within that 10% margin. In contrast, for deterministic gradient descent (DGD) methods, fewer than 10% of optimizations land within 10% of the best result.

We have also included a table demonstrating that although the SGD-based approach is a local optimization method, it trades off global optimality for orders-of-magnitude lower computational requirements.

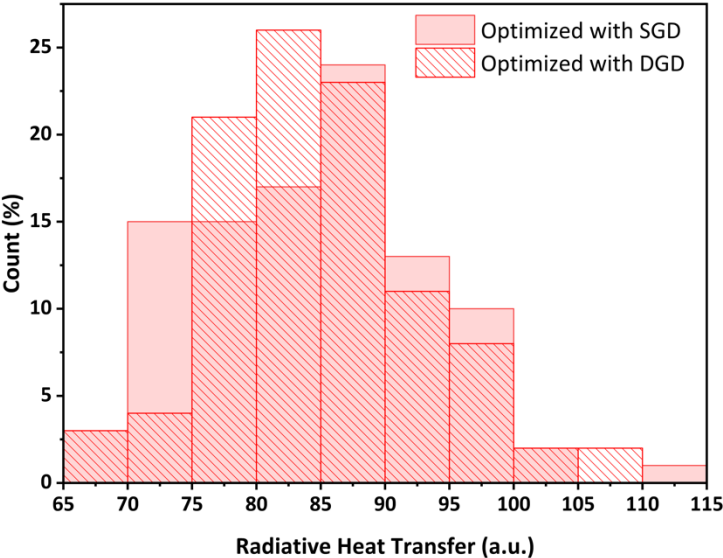


Fig. S4 Comparison of statistical distributions between SGD and DGD methods.

Table. S1 Comparison between different optimization algorithms

Strategy	Global or local optimization?	Advantages	Disadvantages
Particle Swarm ⁷	Global	Good global optimization	Extremely slow (hours to months)
Bayesian optimization ⁸			
Genetic algorithm ⁹			
Gradient descent based ^{10,11}	Local	Efficient optimizations (minutes) and high precision compared with deep learning approach	Chances to stuck at local minima
Deep learning (neuron network) ^{12–14}	Depends on the specific network structure	Extremely quick optimization (seconds or sub-seconds)	Data hungry and challenges in generalizability
Forward design (intuition and parameter sweep)	Not applicable	Clear physics understanding	Does not work for high-dimensional optimizations

5. *The authors cite the need for non-metallic radiative suppression in electronic thermal protection and thermoelectric systems. It would be interesting to briefly discuss how this spectral misalignment strategy used in near-field thermophotovoltaics for waste heat recovery (see, for example, APPLIED PHYSICS LETTERS 107, 091106 (2015)), as both aim to suppress sub-bandgap parasitic heat transfer. Would the same design rational presented here applicable to the near-field heat transfer as well? What would be different?*

Response: We thank the reviewer for this insightful comment and agree that there is a conceptual connection to near-field thermophotovoltaics (NF-TPV), where spectral selectivity and spectral mismatch are used to suppress sub-bandgap parasitic transfer while maintaining above-bandgap photon flux. We will cite the suggested work (Appl. Phys. Lett. 107, 091106 (2015)) and add a brief discussion to clarify similarities and key differences.

- 1) **The “Overlap optimization” philosophy is a unifying design principle.** At a high level, both our approach and NF-TPV filtering strategies can be viewed as optimizing the spectral overlap between emitter and absorber spectra. In our case, the goal is to minimize the overlap between the emissivities of the two facing surfaces over the relevant thermal band. In NF-TPV, the goal is to minimize the near-field photon transmission in the PV’s sub-bandgap region. Thus, the underlying design rationale—engineering dispersive optical responses to reduce unwanted spectral overlap—is indeed shared across both far-field and near-field heat transfer scenarios.
- 2) What fundamentally distinguishes the near field is its dependence on in-plane wavevector and evanescent channels. Near-field heat transfer is dominated by evanescent modes with large in-plane wavevector $k_{\parallel}$ (i.e., $k_{\parallel} \gg k_0$), and the exchange is governed by a mode-resolved transmission function $\xi(\omega, k_{\parallel})$ rather than solely by far-field emissivity $\varepsilon(\omega)$. In fluctuational electrodynamics, the heat flux involves an integral over both ω and $k_{\parallel}$, incorporating both propagating and evanescent channels. As a result, a design optimized for far-field propagating—such as the Bragg reflectors constructed in this work—cannot be directly applied to the near field. Instead, the design objective must be reformulated to minimize the $(\omega, k_{\parallel})$ -resolved transmission within the near-field heat transfer optimization framework.
- 3) In the near field, radiative heat transfer is often enhanced by surface modes (plasmon/phonon polaritons) and high- $k_{\parallel}$ tunneling. Suppression therefore requires controlling not only the spectral position but also the surface-mode density and coupling strength across $k_{\parallel}$. **Current studies on suppressing near-field radiative heat transfer mainly focus on selecting materials or coatings with minimal surface modes and reducing photon tunneling^{15–17}.** The general optimization framework employed in this

work has not yet been leveraged for near-field heat transfer suppression and could therefore be of great interest for future investigations.

In the manuscript, we have added this brief discussion at the end. “The same philosophy can be extended to near-field applications^[63,64], where heat exchange is dominated by evanescent modes with large in-plane wavevectors. In that regime, the design objective must be reformulated to minimize the mode-resolved transmission function—rather than far-field emissivity alone—with careful consideration of surface-mode coupling.”

Reviewer #3 (Remarks to the Author):

In the present work the authors propose a broadband radiative heat transfer suppression scheme based on two dielectric metasurfaces made of distributed Bragg reflectors. The layer thicknesses of the two stacks are optimized using gradient descent with the goal of reducing radiative exchange between them. Essentially, what the authors are aiming to achieve is to instead of trying to make each surface weakly emitting across the entire infrared band, they design two surfaces that are spectrally incompatible with each other, so that frequencies emitted efficiently by one terminal are, as much as possible, not absorbed efficiently by the other. This reduces the spectral overlap that normally enables radiative heat transfer. This is then verified experimentally.

The overall idea is interesting and potentially useful, especially for applications where metallic coatings are not desirable but suppression of radiative heat transfer is still needed. The use of metasurface based multilayer coatings and the optimization of the two interacting terminals rather than a single surface also represent an interesting design strategy in my opinion. The work therefore points toward an interesting and novel direction for controlling radiative exchange using engineered photonic structures.

However there are several points that should be clarified or strengthened.

- 1. My main remark concerns how the paper frames the work relative to bandwidth limits and the discussion of Bode Fano and Rozanov bounds. The manuscript refers to the idea of "going beyond bandwidth constraints" and mentions "circumventing the Bode Fano bound because it applies to continuous bandwidths". However the device considered here is still linear time invariant and passive. As such its scattering response must still obey the usual constraints. Although I am aware that the authors do not explicitly claim to "break these bounds", the current wording may easily lead readers to interpret the results in that way. In addition the manuscript does not provide a quantitative assessment connecting the device performance to those bounds. The discussion therefore remains largely qualitative and speculative. It would be helpful if the authors clarified more precisely what they mean by circumventing bandwidth limits in this context. For instance, the thermal spectrum being integrated in the heat transfer calculation is still "continuous", so the statement that the approach avoids "continuous bandwidth" constraints is not immediately clear.*

Response: We thank the reviewer for the positive comments and thoughtful suggestions. Yes, our structures are passive, linear, and time-invariant; we do not claim to violate these bounds. Our approach instead targets a different objective: minimizing the pairwise spectral overlap that governs radiative exchange between two bodies. By co-designing an aperiodic dielectric reflector pair with complementary dispersion, we distribute stop/pass

behavior across the thermal band to reduce the integrated overlap under a small thickness and fabrication budget, rather than attempting to realize a single-surface extreme response within a single stopband, which according to Bode–Fano/Rozanov trade-offs imposes bandwidth and footprint constraints. Here, we circumvent the bandwidth limits by engineering the spectral piecewise mismatch between two surfaces over the entire thermal band rather than focusing on the bandwidth performance of either surface alone. To avoid misinterpretation, we have revised the entire manuscript to remove ambiguous phrasing such as "circumventing the Bode–Fano bound" and "avoid continuous bandwidth," and instead state clearly that our contribution lies in forming distributed complementary stop/pass features between the two surfaces to reduce the integrated overlap.

Finally, to ground the discussion quantitatively, we have added a comparison against standard periodic DBR baselines with the same layer count in the Supporting Materials, Section 8, reporting the resulting integrated suppression metric as shown below as well. The contribution is therefore framed as achieving improved suppression within a small footprint without mediating performance-bandwidth-thickness trade-offs.

In the manuscript, we have added: “To further quantify this comparison, Supplementary Materials, Section 8 presents a benchmark between the optimized 7-L design and conventional periodic DBRs with the same number of layers. Results show that, based on the calculated emissivity spectra, the 7-L aperiodic DBR pair achieves a reduction of more than 0.1 in mutual emissivity relative to the analytical periodic design within the target frequency range. These results indicate that, even in the presence of strongly dispersive and absorptive materials, the optimization strategy employed here remains effective, outperforming traditional analytical designs by enabling greater control over the spectral response and convergence toward improved solutions.”

In the SM, we added:

Section 8: Quantitative comparison with periodic DBRs

To enable a quantitative comparison with a conventional periodic DBR, we consider a structure composed of the same materials and number of layers as used in this work: alternating high-index amorphous Si and low-index SiO₂ layers, with 7 layers in total (3 pairs plus an additional high-index layer on top), on a SiO₂ substrate. For the analytical DBR design, the layer thicknesses satisfy $n_H t_H = n_L t_L = \frac{\lambda_0}{4}$, where λ_0 is the center wavelength, also referred to as the Bragg wavelength. For simplicity, we assume nondispersive refractive indices of $n_H = 3.45$ for Si and $n_L = 1.52$ for SiO₂.

Within the design range of 500–3000 cm⁻¹, we consider two periodic DBR designs with Bragg wavelengths of 6 μm and 9 μm, respectively, so that their stopbands are offset

from each other. These wavelengths approximately correspond to the peak blackbody wavelengths at 500 K and 300 K, respectively, according to Wien's displacement law. Fig. S25 shows the calculated emissivity spectra of these periodic DBR structures, together with the corresponding mutual emissivity, compared with the optimized 7-L design presented in this work.

One important point is that the 9 μm range overlaps with the Reststrahlen band of SiO_2 , where SiO_2 exhibits strong phonon-polaritonic and dispersive behavior. As a result, emission suppression in this spectral region is intrinsically difficult, consistent with the behavior observed in our optimized design as well. Overall, the optimized 7-L aperiodic DBR structure outperforms these simple analytical periodic DBR designs under the same conditions, exhibiting a reduction of more than 0.1 in mutual emissivity within the designed frequency range.

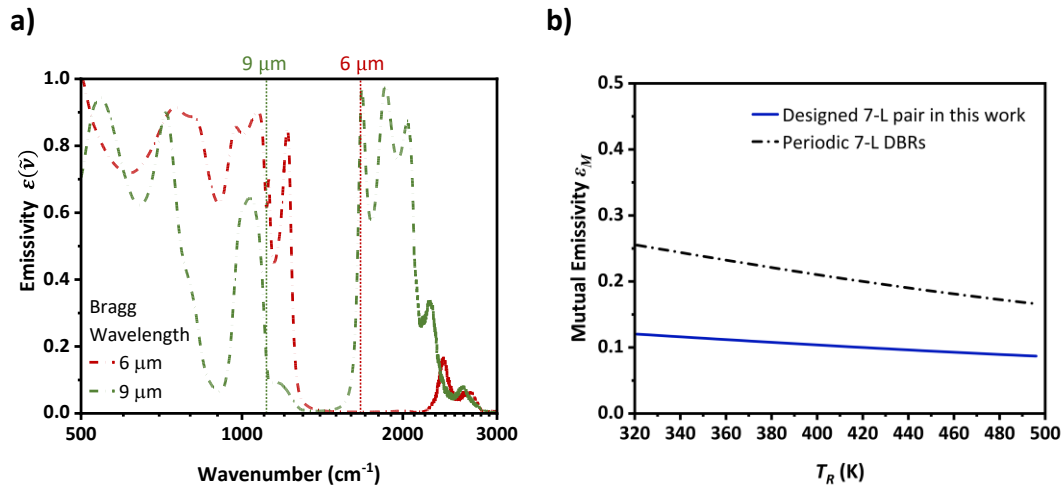


Fig. S22 (a) Emissivity spectra of traditional periodic DBRs with Bragg wavelengths of 6 μm and 9 μm . (b) Comparison of calculated mutual emissivity between the designed 7-L DBRs and periodic DBRs.

2. A second important point is about the consistency between the modeling and the experimentally measured heat transfer performance. The manuscript states suppression levels inferred from emissivity spectra as well as suppression levels obtained from direct heat transfer measurements. However the two estimates differ noticeably, especially for the stronger performing multilayer design. The authors suggest possible explanations such as spectral contributions outside the original design window, but at present the central performance metric is not predicted consistently by the modeling framework used during the inverse design stage. To me, this leaves the impression that the optimization target is a "proxy" for the actual measured quantity rather than a direct prediction of it. I think

strengthening the link between the design objective and the experimentally measured heat transfer would make the central claim more convincing.

Response: We thank the reviewer for this thoughtful comment. We agree that the link between the design objectives and the experimental measurement of heat transfer is not sufficiently highlighted and that the logical flow is missing.

In fact, the initial design had to accommodate unavoidable material uncertainties, along with assumptions and approximations that were later found to adversely affect the results: (A) the material properties at longer wavelengths are challenging to characterize; (B) long-wavelength emissivity measurements are beyond the capabilities of our facilities; (C) based on a quick Planck distribution estimate, the emission outside the 500–3000 cm^{-1} range contributes relatively little; and (D) in the stochastic gradient descent (SGD) procedure, accounting for angle-dependent emissivities is computationally expensive.

Given the lack of complete material properties, limited computational resources, and restrictions on equipment capabilities, the design was completed as described, and the corresponding emissivity measurement serves as an illustration of that design. Subsequent direct radiative power measurements revealed discrepancies between the design and the measured performance. To explain these discrepancies, we performed indirect absorptivity measurements covering the long-wavelength range and large angles. Now, with a complete picture, the devices can be optimized by selecting appropriate materials, incorporating long-wavelength and angle-dependent emissivities into the modeling.

Accordingly, we have added a paragraph at the end of the manuscript to outline the full optimization pathway for achieving enhanced radiative suppression performance with compact all-dielectric platforms: “A systematic analysis was conducted to investigate the discrepancy between direct measurements and indirect estimations. Full details, including the roles of the wavelength range, angular dispersion, and partial transparency, are provided in Supplementary Materials, Section 7. In brief, two primary factors were identified: omission of spectral contributions below 500 cm^{-1} and the assumption of diffuse emission (i.e., angle-independent emissivity) in the original design. To validate these findings, cross-validation was performed using an FTIR spectrometer covering an extended spectral range of 30–8000 cm^{-1} . Rather than directly measuring emissivity, absorptivity was determined as $\alpha = 1 - R - T$, with the influence of partial transparency verified to be negligible. Based on the extended-range absorptivity measured at 10° (normal incidence was not achievable due to setup limitations), the calculated mutual emissivity (dash-dot lines) shows significantly improved agreement with the direct radiometer measurements, particularly for the silica substrate and the 3-L DBR pair (Fig. 4(d)). To further account for large-angle contributions, absorptivity was also characterized

at a polar angle of 75°, and a two-angle averaged effective emissivity was constructed using data at 10° and 75°. This approach yields additional improvement, with the mutual emissivity of the paired structure (dotted lines) showing closer agreement with the direct measurements (Fig. 4(e)). Overall, this analysis resolves the discrepancy between direct and indirect methods and demonstrates that incorporating both far-IR spectral contributions and angular dependence is essential—and sufficient—for accurate performance prediction and reliable suppression design. Despite these corrections, both the 3-L and 7-L metasurface pairs exhibit substantial radiative crosstalk reduction compared to the fused silica baseline within the operating temperature range, thereby validating the proof of concept.”

3. *Related to this, part of the modeling framework relies on treating the structures as effectively opaque when extracting emissivity from reflection measurements. However the manuscript also notes that this assumption breaks down in certain spectral regions where transmission becomes non negligible. When the structures are not strictly opaque, the relationship between reflection and emissivity becomes more complicated and this can affect both the emissivity spectra used for the analysis and the calculation of mutual emissivity between the two terminals. Since the suppression mechanism relies on carefully shaping the spectral response, such modeling assumptions could influence the predicted heat transfer suppression. I suggest that the authors should address this point more carefully in the modeling and design procedure would improve the internal consistency of the analysis.*

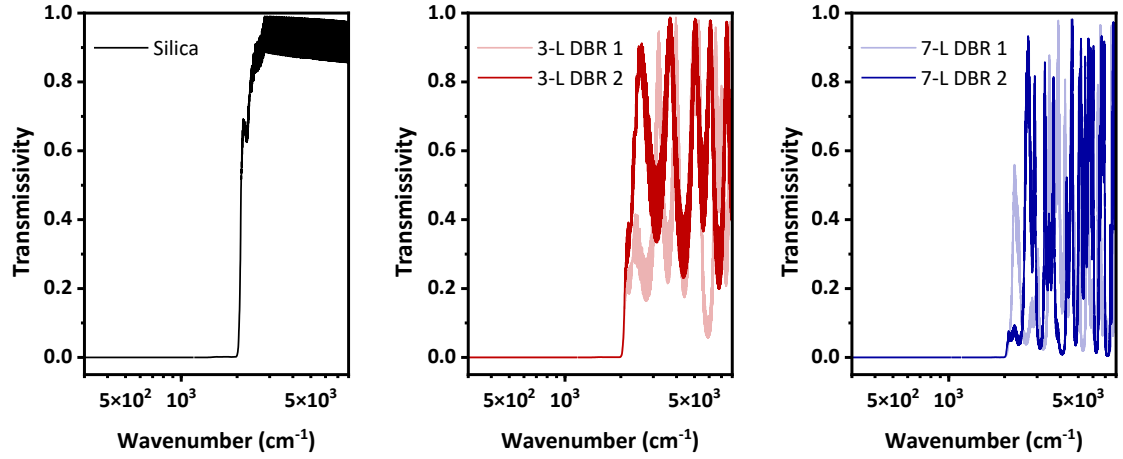
Response: Thank you for this thoughtful comment. We agree that accounting for transparency in the relevant spectral range would lead to a more rigorous and refined analysis.

However, its influence on the performance and on the design is small. We have added Section 7.2 in the SM to discuss the effect of non-opaqueness which is also attached here:

Section 7.2 Effect of non-opaqueness

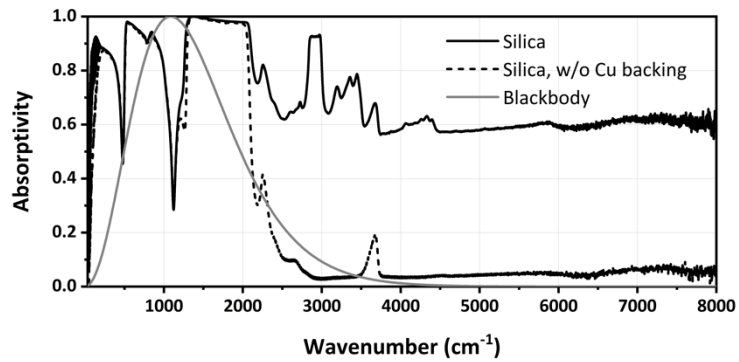
To illustrate that the opaque assumption introduces only minor discrepancies, we first calculated the transmissivity of the fused silica substrate and the designed 3-L and 7-L structures. The transmissivity is nonzero only at wavenumbers $>2000 \text{ cm}^{-1}$ ($5 \mu\text{m}$). According to the following Eq. (S16), thermal emission in this range accounts for only 1.3% of total blackbody emission at 300 K and 16.1% at 500 K.

$$\eta = \frac{\int_0^{5\mu\text{m}} E_{b,\lambda}(\lambda, T) d\lambda}{\sigma T^4} \quad (\text{S16})$$

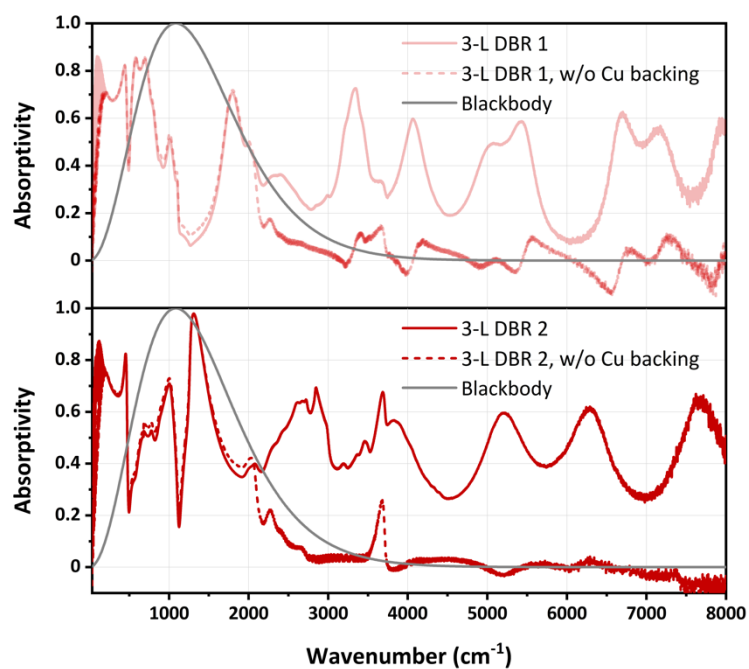


[Fig. S17 Calculated transmissivity of silica, designed 3-L pair, and 7-L pair.](#)

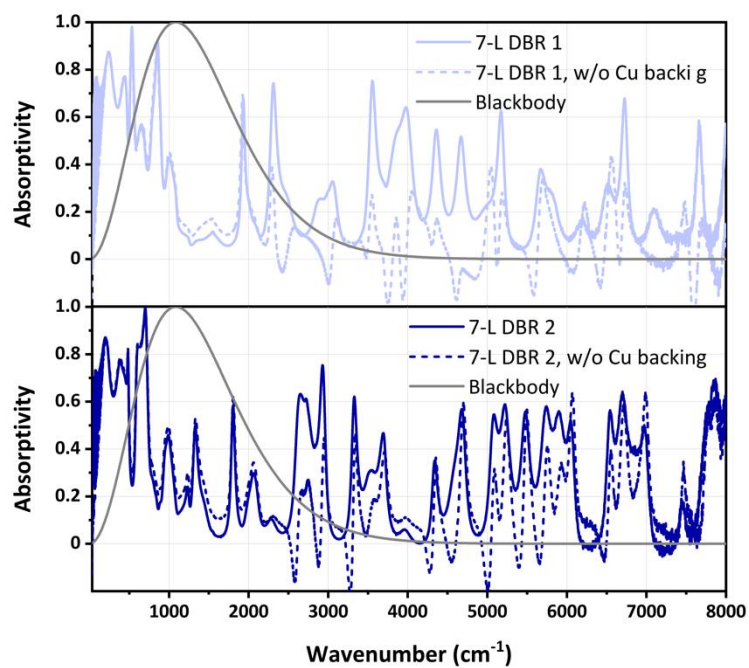
Moreover, in [Figs. S18–S20](#) we present the measured absorptivity using the long-wavelength-range FTIR spectrometer with and without Cu backing (i.e., where the transmissivity is non-zero). The blackbody radiation weight corresponding to the difference between 500 K and 300 K is also plotted to visually demonstrate that most of the discrepancy from the Cu-backed results lies outside the blackbody emission band and thus does not contribute to the main performance. Based on these data, the calculated mutual emissivity, both with and without considering transmission through the designed devices, is shown in [Fig. S21](#). From this, we can further conclude that, at relatively low operating temperatures, the influence of omitting device transmission and assuming full opacity remains relatively small.



[Fig. S18 Absorptivity spectra of silica with and without Cu backing.](#)



[Fig. S19 Absorptivity spectra of 3-L DBR pair with and without Cu backing.](#)



[Fig. S20 Absorptivity spectra of 7-L DBR pair with and without Cu backing.](#)

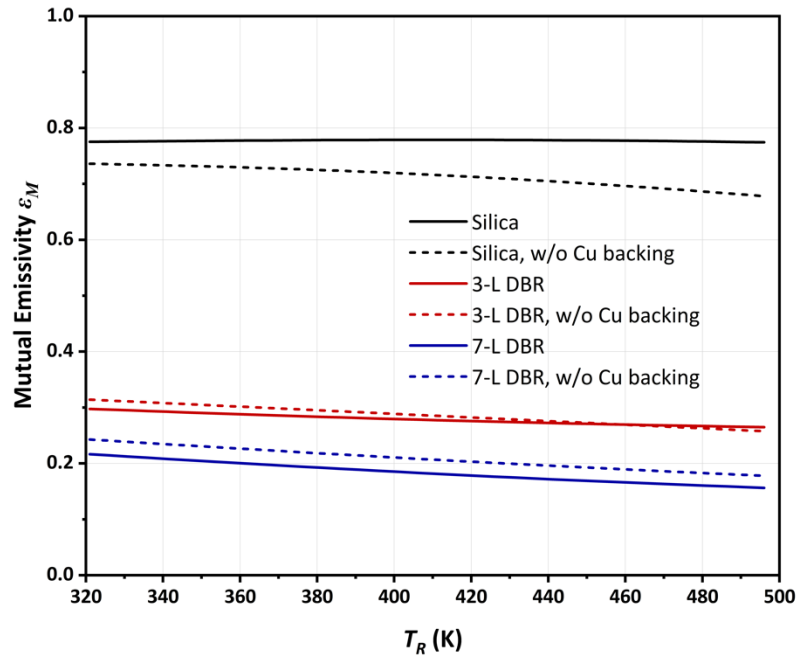


Fig. S21 Calculated mutual emissivity of silica, 3-L DBR pair, and 7-L DBR pair based on long-wavelength-range absorptivity with and without Cu backing.

4. *This is somewhat more speculative, but it may be useful to consider possible alternative approaches that could achieve a similar effect with simpler design procedures. For instance, one could also use two chirped reflectors designed to provide broadband reflection, but with slightly different reflection or emissivity profiles across the spectrum. Here the suppression of radiative exchange would arise from a broad spectral mismatch between the two surfaces, rather than from a "piecewise" mismatch created by multiple narrow resonances. Such a strategy might be significantly easier to design since chirped multilayer reflectors are well understood and can often be obtained using straightforward analytical design rules without the need for optimization. Also, designs that rely less on narrow resonances and resonance alignment may be more robust to fabrication imperfections or layer thickness variations. Perhaps it would be helpful if the authors clarified how the present approach compares with such alternatives. In particular, is the main advantage of the proposed design that it can achieve the desired suppression with fewer layers or reduced total thickness, or that it offers a higher degree of control over the spectral response?*

Response: We thank the reviewer for this thoughtful suggestion. We agree that chirped multilayer reflectors serve as a natural baseline for broadband reflection and could, in principle, be paired with a second reflector having a slightly different spectral profile to reduce radiative exchange through a broad spectral mismatch. However, in the context of

this work, our proposed approach aims to establish a **general optimization framework**, rather than focusing on a specific structural design. Its broader applicability beyond chirped reflector configurations is supported by the following two points:

- 1) **From a design framework perspective**, our method employs end-to-end optimization that intrinsically accounts for material dispersion and absorption, enabling a high degree of design flexibility and effective performance optimization. As demonstrated in the Response to Comment 1, even in the presence of strongly dispersive and absorptive materials, the optimization remains efficient and can outperform traditional analytical DBR designs by providing enhanced control over the spectral response and enabling convergence toward improved solutions.
- 2) **From the standpoint of specific broadband chirped reflector implementations**, they can be significantly affected by material dispersion and absorption, which degrade their reflective performance and often require more complex designs or numerical optimization to compensate¹⁸. In idealized cases with low-loss materials, chirped reflectors designed for central wavelengths between 5 and 10 μm typically require average layer thicknesses on the order of 0.8–1.7 μm and total layer counts exceeding $10^{19,20}$. While such designs can achieve broadband reflection, they come at the cost of substantially increased thickness and fabrication complexity. In the context of radiative heat transfer suppression, maximizing broadband reflectivity is not the primary objective; instead, the key metric is the **mutual radiative exchange between two surfaces integrated over the thermal spectrum**. Pursuing extreme broadband performance may therefore lead to unnecessary complexity without proportional benefit. When accounting for material loss, dispersion, and practical fabrication constraints, straightforward analytical design rules for chirped reflectors become insufficient, highlighting the need for a more general optimization approach.

In summary, this work provides a broadly applicable design framework that enhances flexibility while keeping fabrication complexity manageable—without being tied to a specific structure. The same optimization procedure can be readily applied to chirped reflector configurations to further improve their performance.

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