

Thin-Film Radiative Thermal Diode with Large Rectification

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We propose a mechanism to substantially rectify radiative heat flow by having one side of the thermal diode offering a large contrast in the local density of electromagnetic states, while the opposite side serving as a narrow bandpass filter that limits the transport to a desired spectral range. To demonstrate, we realize such a scenario by matching thin films of metal-to-insulator transition materials and polar dielectrics in the near field. By leveraging the distinct scaling behaviors of the local density of states with film thickness for metals and insulators, we theoretically achieve rectification ratios over 140, a 10-fold improvement over the state of the art, with nanofilms of vanadium dioxide and cubic boron nitride in the parallel-plane geometry at experimentally feasible gap sizes (~ 100 nm). Our rational design offers relative ease of fabrication, flexible choice of materials, and robustness against deviations from optimal film thicknesses. We expect this work to facilitate the application of thermal diodes in solid-state thermal circuits and energy-conversion devices.

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I. INTRODUCTION

Thermal diodes, in analogy to electrical diodes, are two-terminal devices that rectify the flow of heat and are desired in many applications, including thermal management, energy conversion, and thermal circuits and computing [1–3]. The key figure of merit for a thermal diode is its rectification ratio, defined as $R = (Q_F - Q_R)/Q_R$, where Q_F denotes the larger heat current under a forward temperature bias and Q_R is the smaller reverse current [4]. A 10-fold rectification is often necessary for technical efficiency. However, despite extensive studies of thermal diodes mediated by phonons, electrons, and photons, most experimentally demonstrated rectification ratios remain around one or much smaller [4–8]. With potential for orders-of-magnitude rectification over a wide range of temperatures, near-field radiative thermal diodes (NFRTDs), leveraging the large electromagnetic local density of states (LDOS) close to a surface, have recently gained broad interest [7,9–21].

Research on NFRTDs began with bulk materials in the parallel-plane geometry separated by a vacuum gap. By

using the temperature-dependent surface phonon polaritons (SPhPs) in two polymorphs of silicon carbide (SiC), Otey *et al.* [9] obtained a rectification ratio of 0.41 at a gap size of 100 nm. Subsequently, the surface modes supported by doped semiconductors [10,18], phase-change materials [11,12,15], polar dielectrics [14,17], and two-dimensional materials [16,20] were widely explored, together with nanoscale structural innovations (e.g., multilayers and linear gratings), leading sometimes to rectifications over 10. By using nanospheres instead of parallel planes, the largest rectification, to date ($> 10^4$), has been theoretically predicted [13,19]. However, experiments on NFRTDs are rare and have only reported rectifications around one [7], largely due to challenges in achieving 10-nm gaps between parallel planes and in characterizing radiative thermal transport between delicate nanostructures.

Here, we propose that large rectification of radiative heat flow can be readily achieved by matching and optimizing a thin film of metal-to-insulator transition (MIT) materials and another of polar dielectrics in the parallel-plane geometry, which is the most technologically promising, at experimentally feasible gap sizes [Fig. 1(a)]. We begin with an asymptotic analysis of hypothetical materials to illustrate the physical mechanism, followed by rigorous calculations for thermal diodes composed of realistic materials and structures. We show over 140-fold rectification

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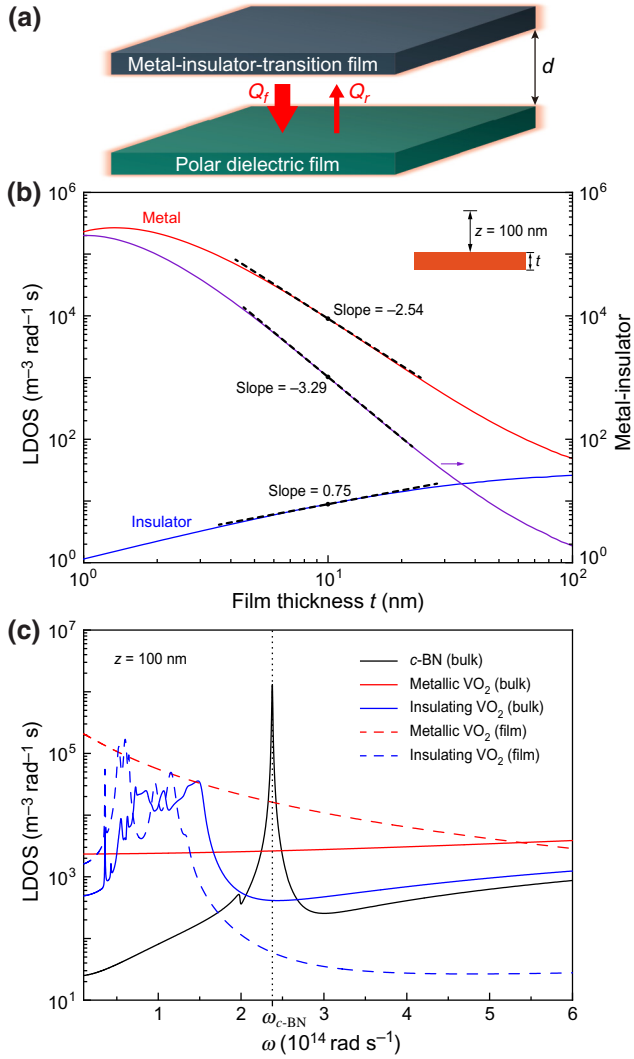


FIG. 1. (a) Schematic of the thin-film radiative thermal diode. (b) LDOS of p -polarized evanescent modes 100 nm above a metal and an insulator film and their ratio at $\omega = 10^{14}$ rad/s with varying film thickness. For the metal, we assume $\omega_p = 10\omega$, so $\varepsilon'_m \approx 1 - \omega_p^2/\omega^2 = -99$, and set $\varepsilon''_m = 10^{-3}$ to represent a low loss. For the insulator, we set $\varepsilon_i = 2 + 10^{-3}i$. (c) LDOS above c -BN and VO_2 .

near room temperature at around 100-nm gaps, together with a high robustness against deviations from optimal film thicknesses. Our results open up the possibility for realizing orders-of-magnitude thermal rectification using state-of-the-art experimental techniques [22–25].

II. RESULTS AND DISCUSSION

To design a high-performance NFRTD, we first focus on the near-field LDOS because a large LDOS usually leads to a large radiative heat flow and vice versa [26]. Our idea is to create a scenario where one side of the diode offers a substantial LDOS contrast as the temperature varies, while

the other provides a sharp LDOS peak which serves as a narrow bandpass filter and limits the heat flow to a desired frequency range for ease of control. Such a scenario can be established, for example (Fig. 1), by exploring the phase transitions of MIT materials [27] in conjunction with the SPhPs supported by polar dielectrics [28–30].

The key to maximizing rectification is to maximize the contrast of near-field LDOS across the metal-to-insulator transition. We achieve this by simply using a MIT thin film instead of a bulk, because the low-frequency LDOS of a metal film increases substantially with decreasing film thickness while the opposite often happens for an insulator film, as shown in Figs. 1(b) and 1(c). Intuitively, as the thickness of a metal film becomes thinner than the skin depth, the surface plasmon polaritons (SPPs) at the two interfaces couple within the film and split into two branches of resonant modes—the low-frequency symmetric and the high-frequency antisymmetric modes [31,32]. Compared to the SPPs on a bulk material, the symmetric modes of a thin film shift to higher parallel wavevectors and lead to enhanced LDOS that are thermally accessible [32]. On the contrary, the LDOS for an insulator may decrease over a wide frequency range due to reduced emitting volume as the film thickness decreases [33].

To quantify the effect of film thickness on the LDOS contrast, we derive approximate expressions in the near field of both metals and insulators, assuming nonmagnetic materials so only p -polarized surface modes exist. For p -polarized evanescent waves, the LDOS at a distance z above a suspended film of thickness t is given by [26]

$$\begin{aligned} \rho(\omega, z) &= \int_{k_0}^{\infty} \rho_w(\kappa, \omega, z) d\kappa \\ &= \int_{k_0}^{\infty} \frac{\kappa^3}{2\pi^2 \omega |\gamma_0|} e^{-2\text{Im}(\gamma_0)z} \text{Im}(R_f) d\kappa, \end{aligned} \quad (1)$$

where ρ_w denotes the wavevector-resolved local density of states (WLDOS) [34,35], κ is the parallel wavevector, $k_0 = \omega/c$, $\gamma_0 = \sqrt{k_0^2 - \kappa^2}$, and R_f is the reflection coefficient of the film. Since the near-field heat flow is often dominated by high- κ modes with $\kappa \gg k_0$, the WLDOS can be simplified as

$$\rho_w(\kappa, \omega, z) \approx \frac{1}{2\pi^2 \omega} \kappa^2 e^{-2\kappa z} \text{Im}(R_f). \quad (2)$$

We first consider a film of Drude metal with plasma frequency ω_p and negligible loss, the permittivity of which can be written as $\varepsilon_m = \varepsilon'_m + i\varepsilon''_m$, where $\varepsilon'_m \approx 1 - \omega_p^2/\omega^2$ and $\varepsilon''_m \approx 0$. By solving for the poles of R_f at sufficiently low frequencies ($\omega \ll \omega_p$), we find the symmetric resonant modes characterized by $\kappa_r = 2/(|\varepsilon'_m|t)$ (see the Supplemental Material for details [36]). The increase of κ_r with decreasing film thickness reflects stronger coupling

of SPPs within thinner films [31,32]. Upon resonance we have $\text{Im}(R_f) = |\varepsilon'_m|/\varepsilon''_m$, so the WLDOS is written as

$$\rho_{w,m}(\kappa_r, \omega, z) = \frac{2}{\pi^2 \omega \varepsilon''_m |\varepsilon'_m|} e^{-4z/(|\varepsilon'_m|t)} t^{-2}, \quad (3)$$

with a full width at half maximum of $\Delta\kappa = (4\varepsilon''_m/\varepsilon'^2_m)t^{-1}$. The LDOS of a metal film can then be approximated by

$$\rho_m \approx \rho_{w,m}(\kappa_r, \omega, z) \Delta\kappa = \frac{8}{\pi^2 \omega |\varepsilon'_m|^3} e^{-4z/(|\varepsilon'_m|t)} t^{-3}, \quad (4)$$

which increases with decreasing film thickness roughly as t^{-3} and peaks at $t = 4z/(3|\varepsilon'_m|)$. Likewise, for an insulator film characterized by $\varepsilon_i = \varepsilon'_i + i\varepsilon''_i$, with low loss and no surface modes, the low-frequency LDOS is derived as [36]

$$\rho_i \approx \frac{27\varepsilon''_i(\varepsilon'^2_i + 1)}{16e^3 \pi^2 \omega \varepsilon'^2_i} z^{-4} t. \quad (5)$$

Here, the linear scaling with film thickness reflects the linearly decreasing volume. Equations (4) and (5) show that for a hypothetical MIT film, the LDOS contrast between the metal and the insulator phase could potentially scale as t^{-4} , in consistency with our intuitive pictures. Further, we plot in Fig. 1(b) rigorously calculated results which agree well with the asymptotic analysis.

Guided by the physical insights from ideal materials, we now explore the potential of real MIT materials for large thermal rectification. To this end, we compute the near-field LDOS for vanadium dioxide (VO₂)—a prototypical MIT material with a phase-transition temperature around 341 K [37–39]. As expected, the LDOS of a 1-nm-thick VO₂ film [38] is up to two orders-of-magnitude larger than that of bulk VO₂ in the metal phase over a wide spectral range, while a similarly dramatic LDOS decrease is observed in the insulator phase [Fig. 1(c)]. This leads to a substantial increase in the LDOS contrast and forms the basis for large rectification, despite the fact that VO₂ has non-negligible loss in both phases and fails to quantitatively follow the asymptotic relation with film thickness (Fig. S1) [36]. To form a thermal diode [Fig. 1(a)], we choose cubic boron nitride (*c*-BN) [40,41] as the polar dielectric that pairs with VO₂, because it supports strong SPPs around $\omega_{c\text{-BN}} = 2.376 \times 10^{14}$ rad/s, where VO₂ shows a large metal-to-insulator LDOS ratio [Fig. 1(c)].

With the materials of our NFRTDs selected, we proceed to exact calculations of the forward and reverse heat flows in the parallel-plane configuration. Based on the theoretical framework of fluctuational electrodynamics, the heat flux between two closely spaced bodies across a vacuum gap is

given by [42,43]

$$q(T_1, T_2, d) = \int_0^\infty \frac{d\omega}{4\pi^2} [\Theta(\omega, T_1) - \Theta(\omega, T_2)] \times \int_0^\infty d\kappa \kappa [\tau_s^{12}(\omega, \kappa) + \tau_p^{12}(\omega, \kappa)], \quad (6)$$

where $\Theta(\omega, T) = \hbar\omega/[\exp(\hbar\omega/k_B T) - 1]$ is the mean energy of a harmonic oscillator less the zero-point contribution at absolute temperature T ; d is the gap size; and $\tau_s^{12}(\omega, \kappa)$ and $\tau_p^{12}(\omega, \kappa)$ are, respectively, the transmission probabilities for the *s*- and *p*-polarized modes, which can be expressed in terms of the Fresnel reflection coefficients (r) as follows:

$$\tau_{\alpha=s,p}^{12}(\omega, \kappa) = \begin{cases} \frac{(1-|r_\alpha|^2)(1-|r_\alpha^2|^2)}{|1-r_\alpha^1 r_\alpha^2 \exp(2i\gamma_0 d)|^2}, & \kappa \leq k_0, \\ \frac{4\text{Im}(r_\alpha^1)\text{Im}(r_\alpha^2)e^{-2\text{Im}(\gamma_0)d}}{|1-r_\alpha^1 r_\alpha^2 \exp(2i\gamma_0 d)|^2}, & \kappa > k_0. \end{cases} \quad (7)$$

In Eq. (7), $1 - |r_\alpha|^2$ should be replaced with $1 - |r_\alpha|^2 - |t_\alpha|^2$ for suspended thin films, where t_α is the transmission coefficient.

We show in Fig. 2 the transmission probabilities for three diode configurations, namely, bulk VO₂ paired with bulk *c*-BN, VO₂ film (10 nm) paired with bulk *c*-BN, and VO₂ film (10 nm) paired with *c*-BN film (10 nm), at a gap size of 100 nm. The terminal temperatures of the diodes are fixed at $T_{\text{high}} = 351$ K and $T_{\text{low}} = 331$ K to allow complete phase transition of VO₂ [39], which is an isotropic metal at T_{high} in the forward scenario and a uniaxial dielectric when the temperature bias is reversed. Compared with the bulk-bulk case [Figs. 2(a)–2(c)], the film-bulk case [Figs. 2(d)–2(f)] shows a larger forward heat flux dominated by the SPPs within the narrow Reststrahlen band of *c*-BN (Table S1) [36], due to the enhanced low-frequency LDOS of the metallic VO₂ film [Fig. 1(c)]. While in the reverse scenario, a much smaller heat flux is observed because the reduced volume of insulating VO₂ leads to smaller contributions from the broadband nonsurface modes with $\kappa < \text{Re}(\sqrt{\varepsilon_{c\text{-BN}}})k_0$ (left of the *c*-BN light line in Fig. 2). As a result, a much larger rectification ratio (R) of 18.28 is obtained with a VO₂ film and bulk *c*-BN than that of two bulk layers ($R = 0.35$). Moreover, the rectification ratio increases to 24.31 when bulk *c*-BN is replaced with a thin film [Figs. 2(g)–2(i)], which leads to smaller LDOS outside the Reststrahlen band and weaker coupling with the insulating VO₂ film. This further reduces the reverse heat flux, while maintaining a large forward flux. As indicated by the parameter η in Table S1 [36], which represents the heat flux mediated by the SPPs of *c*-BN, creating a scenario characterized by narrowband heat transport is key to achieving large thermal rectification.

By systematically optimizing the film thickness, we predict a maximum rectification of 140.77 by combining

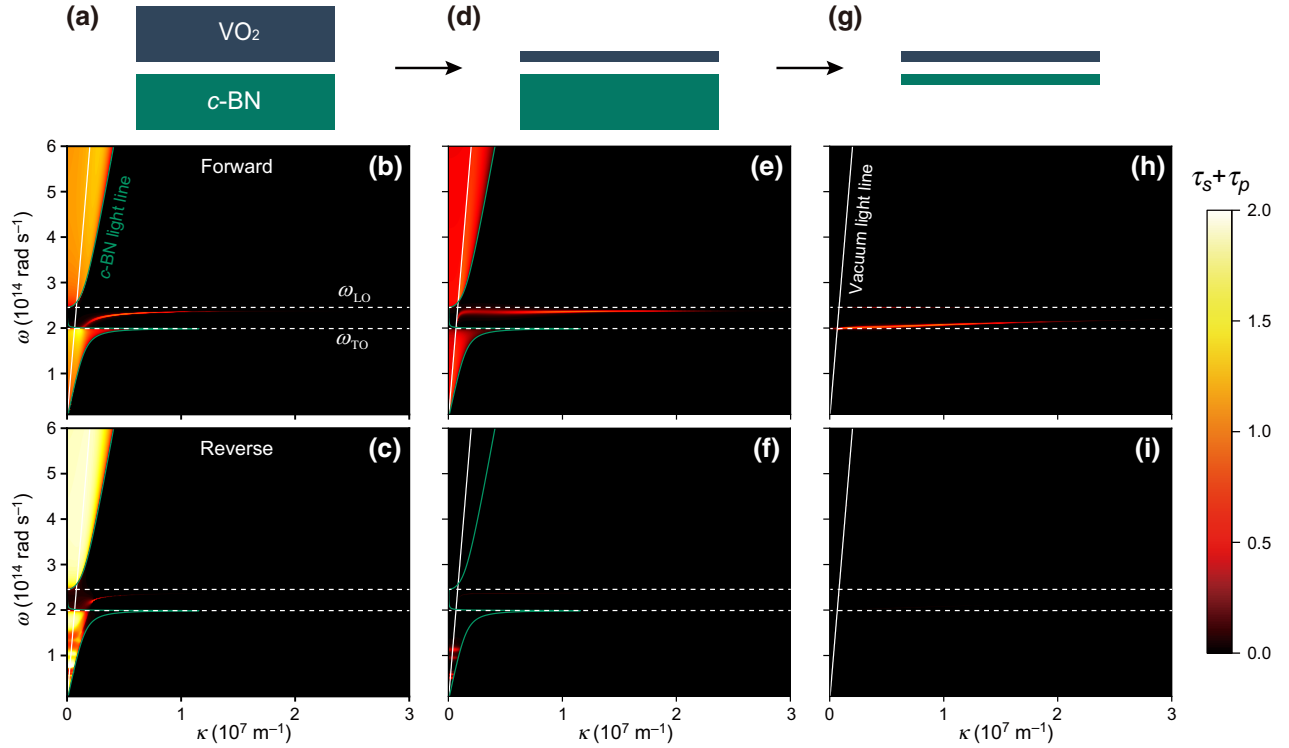


FIG. 2. Comparison of transmission probabilities for bulk and thin-film diodes. (a)–(c) Bulk VO_2 -bulk $c\text{-BN}$, (d)–(f) VO_2 film-bulk $c\text{-BN}$, (g)–(i) VO_2 film- $c\text{-BN}$ film. Vacuum gap is 100 nm wide, and all films are 10 nm thick. Dashed lines mark the zone-center transverse and longitudinal optical phonons (ω_{TO} and ω_{LO}) in $c\text{-BN}$, which enclose the Reststrahlen band.

a 1-nm-thick VO_2 film with a 149-nm-thick $c\text{-BN}$ film (Fig. 3). The dramatic rectification enhancement primarily originates from the ultrathin VO_2 film, which features a much larger metal-to-insulator LDOS ratio around $\omega_{c\text{-BN}}$ [Fig. S2(b)] [36], consistent with our asymptotic analysis. In comparison, the optimal $c\text{-BN}$ film is much thicker. This is because the SPhPs will couple within the $c\text{-BN}$ film if its thickness becomes smaller than the penetration depth ($\approx d = 100$ nm) [30,44,45], leading to a redshift of the LDOS peak to the frequency range where VO_2 shows a smaller LDOS contrast which reduces rectification [Figs. S2(b) and 2(d)] [36]. We also note that, when the VO_2 film is around 1 nm thick, the rectification ratio remains above 90, regardless of the $c\text{-BN}$ film thickness, which again reflects the key role of the ultrathin VO_2 film [Fig. 3 and Fig. S2(c)] [36].

A thermal diode composed of two suspended nanofilms is theoretically appealing but technically challenging. Here, we build upon the optimized suspended-film design and propose a more practical multilayer structure with similarly large rectification ratio (Fig. 4). First, we add to each film a bulk dielectric substrate with a permittivity, ϵ_d , close to that of vacuum. By setting $\epsilon_d = 1 + 10^{-4}i$, we obtain a rectification of 4.40 across a vacuum gap of 100 nm, in contrast to 140.77 for the suspended-film configuration with the same film thicknesses. This is because of the large reverse heat flux dominated by the broadband

propagating modes in the dielectric substrates with $\kappa < \text{Re}(\sqrt{\epsilon_d})k_0$, which can be effectively suppressed by reducing the thickness of the substrates. By using a 1- μm -thick substrate instead of the bulk, we observe essentially the

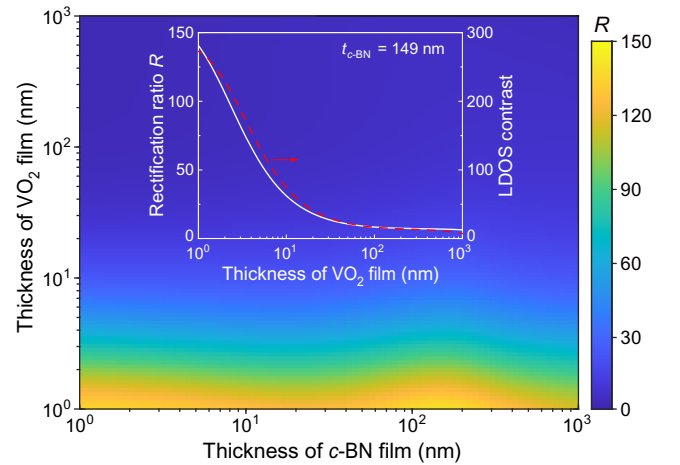


FIG. 3. Rectification ratio of a suspended VO_2 - $c\text{-BN}$ thin-film diode as a function of film thickness at a 100-nm gap. Inset shows rectification and corresponding LDOS contrast at $\omega_{c\text{-BN}}$ with varying VO_2 film thickness, when the $c\text{-BN}$ film thickness is an optimal value of 149 nm.

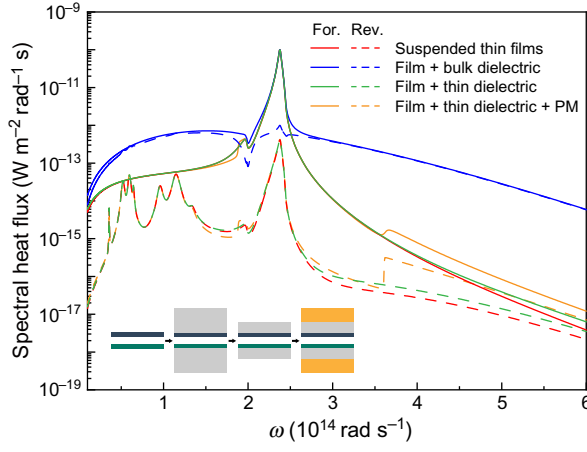


FIG. 4. Spectral heat fluxes of different thin-film diodes in the forward (For.) and reverse (Rev.) scenarios across a 100-nm gap.

same spectral characteristics as those of the suspended-film design and a large rectification ratio of 134.52. We further add a reflective metal layer on the backside of the dielectric substrates to isolate the diode from thermal radiation in the environment. With a perfect metal (PM) [46] as the reflector, the spectrum remains almost unaltered and a rectification ratio of 134.81 is achieved. We note that the dielectric substrate should be thicker than the penetration depth ($\approx d$) of the dominating surface modes ($\kappa d \approx 1$), so that any detrimental effect from the metal reflector is minimized [30,45].

Although the above design is promising, in principle, we have yet to identify real materials that are appropriate to serve as the dielectric substrates and the metal reflectors. The substrates should have vacuumlike permittivities around $\omega_{c\text{-BN}}$ and should not induce new surface modes, which could increase the reverse heat flow and lower the rectification. We find that potassium chloride (KCl) [47], potassium bromide (KBr) [47], silicon (Si) [48], and SiC [26] represent some of the best candidates. As to the backside reflectors, metals with low loss and high reflectivity, such as silver (Ag) and gold (Au), are preferred. In Fig. 5(a), we present the rectification ratios as a function of gap size for a few thin-film NFRTDs consisting of different combinations of the candidate materials. All of the diodes feature rectification ratios over 60 at a gap size of 100 nm (Table S2) [36], with a maximum of 78.85 achieved by the pair of $\text{VO}_2/\text{KBr}/\text{Ag}$ and $c\text{-BN}/\text{SiC}/\text{Ag}$. The rectification ratio of this particular diode further increases to 105.68 at a 50-nm gap, which is readily accessible in the parallel-plane geometry [22,23], and remains greater than 10 even at gap sizes up to about 700 nm. We note that the smaller rectification at larger gaps is due to the rapid decay of the surface modes, while, at smaller gaps, the gap-size-dependent penetration depth of the surface modes is responsible. Indeed, thin films can act like bulk materials in the extreme near

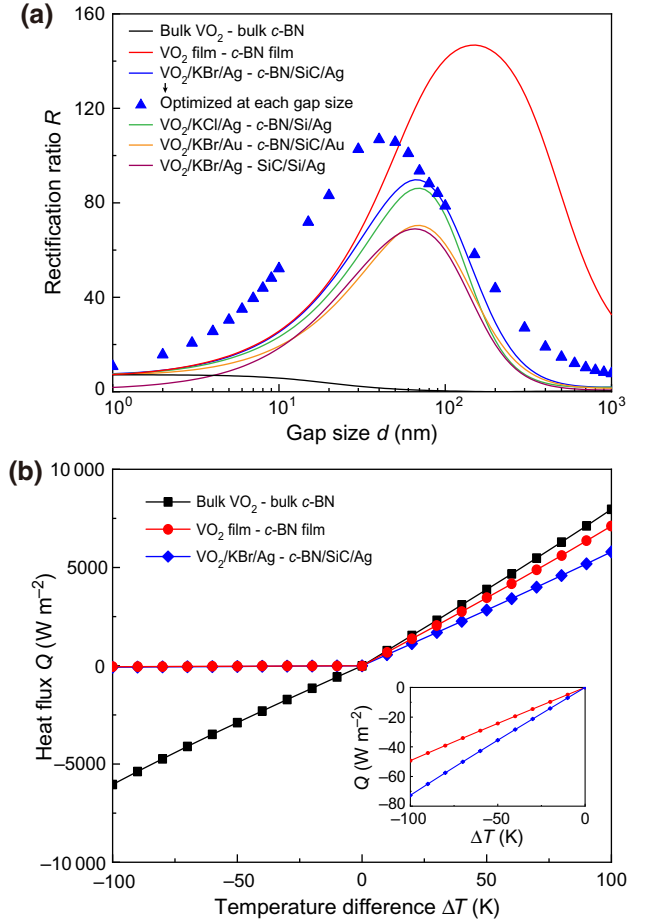


FIG. 5. Performance of optimized thin-film diodes with realistic structures and materials. (a) Gap-dependent rectification ratios. (b) Heat fluxes under different temperature biases at a gap of 100 nm. Terminal temperatures are set as $T_1 = T_{\text{av}} + \Delta T$ and $T_2 = T_{\text{av}} - \Delta T$, respectively, for the VO_2 and the $c\text{-BN}$ sides, with T_{av} fixed at 341 K, which is the nominal phase-transition temperature of VO_2 .

field and results in rectification ratios approaching the bulk-bulk case [30,45,49]. Last but not least, our thin-film diodes are expected to perform well over a wide temperature range [Fig. 5(b)] and against inevitable film thickness variations in fabrication (Fig. S3) [36].

III. CONCLUSIONS

We propose a mechanism to achieve orders-of-magnitude rectification of radiative heat flow between parallel planes across experimentally attainable vacuum gaps. We combine an ultrathin MIT film with a polar dielectric film, so that the latter enables surface-mode-mediated narrowband heat transport over a spectral range where the former provides a large metal-to-insulator LDOS contrast upon phase transition. Our asymptotic analysis reveals that the LDOS ratio could increase with decreasing film thickness (t) as t^{-4} , leading to a similarly dramatic rectification

enhancement. We demonstrate a rectification ratio of over 140 by pairing thin films of VO₂ and *c*-BN at a gap size of 100 nm, which is 400 times better than its bulk counterpart and a 10-fold improvement over state-of-the-art thermal diodes. We further explore realistic multilayer structures and identify a variety of appropriate materials. Our effort paves the way for the long-awaited use of thermal diodes in solid-state energy and logic devices.

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