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Self-similar nanocavity design with ultrasmall mode volume for single-photon nonlinearities

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We propose a photonic crystal nanocavity design with self-similar electromagnetic boundary conditions, achieving ultrasmall mode volume (V_{eff}). The electric energy density of a cavity mode can be maximized in the air or dielectric region, depending on the choice of boundary conditions. We illustrate the design concept with a silicon-air 1D photon crystal cavity that reaches an ultrasmall mode volume of $V_{\mathrm{eff}} \sim 7.01 \times 10^{-5}\lambda^3$ at $\lambda \sim 1550$ nm. We show that the extreme light concentration in our design can enable ultra-strong Kerr nonlinearities, even at the single photon level. These features open new directions in cavity quantum electrodynamics, spectroscopy, and quantum nonlinear optics.

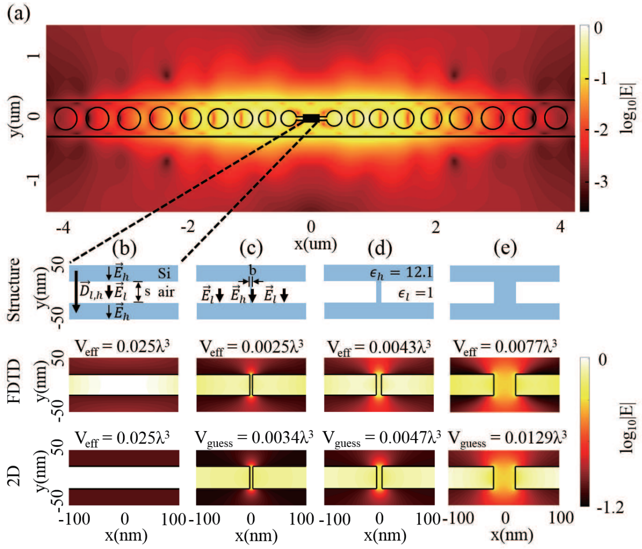

FIG. 1. Cavity field profiles of a slot cavity and slot/bridge cavities with different width of bridges. (a), (b) Slot cavity achieving enhancement with the Type-1 BC ($V_{\mathrm{eff}} = 2.5 \times 10^{-2}\lambda^3$). Here, s denotes slot width, and $s = 40$ nm is used. (c)∼(e) Slot/bridge cavities (SB) achieving enhancement with Type-2 BCs. Top: Index profile of the structure. Here, b denotes bridge width. Middle: 3D FDTD simulation result. Bottom: 2D electrostatic simulation result. (c) $b = 5$ nm narrow bridge ($V_{\mathrm{eff}} = 2.5 \times 10^{-3}\lambda^3$). (d) $b = 10$ nm intermediate bridge ($V_{\mathrm{eff}} = 4.3 \times 10^{-3}\lambda^3$, $V_{\mathrm{guess}} = 3.4 \times 10^{-3}\lambda^3$). (e) $b = 40$ nm wide bridge ($V_{\mathrm{eff}} = 7.7 \times 10^{-3}\lambda^3$, $V_{\mathrm{guess}} = 1.3 \times 10^{-2}\lambda^3$). For the V_{eff} calculation, electric energy density at the middle of the bridge is used as a maximum. This is because corners of the bridges produce a singularity of the field (suppressed by mesh size), but does not affect in typical light-matter interaction. In other words, this is justified by overlap factors, for example, in the Purcell factor.

Optical nanocavities with small mode volume (V_{eff}) and high quality factor (Q) can greatly increase light-matter interaction [1] and have a wide range of applications including nanocavity lasers [2–4], cavity quantum electrodynamics (cQED) [5, 6], single-molecule spectroscopy [7], and nonlinear optics [8–10]. Planar photonic crystal cavities can enable high Q-factors, exceeding 10^6 [11], together with mode volumes that are typically on the order of a qubic wavelength. However, it was shown that by introducing an air-slot into a photonic crystal (PhC) cavity, it is possible to achieve the electromagnetic (EM) mode with small V_{eff} on the order of $0.01\lambda^3$ [12], where λ is the free-space wavelength. This field concentration results from the boundary condition on the normal component of the electric displacement ($\vec{D}$). Here, we propose a method to further reduce V_{eff} by making use of the second EM boundary condition, the conservation of the parallel component of the electric field. Furthermore, these field concentration methods can be concatenated to reduce V_{eff} even further, limited only by practical considerations such as fabrication resolution. The extreme field concentration of our cavity design opens new possibilities in nonlinear optics. In particular, we show that Kerr nonlinearities, which are normally weak, would be substantially enhanced so that even a single photon may shift the cavity resonance by a full linewidth, under realistic assumptions of materials and fabrication tolerances.

The mode volume of a dielectric cavity (described by the spatially varying permittivity $\epsilon(\vec{r})$) is given by the ratio of the total electric energy to the maximum electric energy density [13]:

$$V_{\mathrm{eff}} = \frac{\int \epsilon(\vec{r})|E(\vec{r})|^2 dV}{\max(\epsilon(\vec{r})|E(\vec{r})|^2)}. \tag{1}$$

In typical PhC cavity designs, the minimum cavity mode volume is given by a half-wavelength bounding box, or $V_{\mathrm{eff}} \sim (\lambda/2n)^3$ [14], agreeing with the diffraction limit. However, as is clear from Eq. 1, the mode volume is determined by the electric energy density at the position where it is maximized. Thus, it is not strictly restricted by the diffraction limit. A strong local inhomogeneity in $\epsilon(\vec{r})$ can greatly increase this electric energy density and correspondingly shrink the mode volume.

Figure 1(a) plots the fundamental mode of a silicon-air 1D PhC cavity produced by 3D finite-difference time-domain (FDTD) simulations. This mode pattern (represented here as $|E|$ at the cavity center plane, $z = 0$) is modified only weakly for small perturbations of $\epsilon(\vec{r})$ in the cavity center, and therefore serves to approximate the numerator of Eq. 1. Robinson et al. [12] were able to increase the maximum electric field term in the denominator of Eq. 1 by introducing a thin air slot in the cavity center. This concentration results from the boundary condition on the normal component of the electric displacement (called here the 'Type-1' BC), as illustrated in Fig. 1(b):

$$\epsilon_l E_{l\perp} = D_{l\perp} = D_{h\perp} = \epsilon_h E_{h\perp}, \tag{2}$$

$$E_{l\perp} = \frac{\epsilon_h}{\epsilon_l} E_{h\perp}, \tag{3}$$

where ϵ_l and ϵ_h are the permittivities of the low and high index materials, respectively, and the subscript $\perp$ represents the normal component of the field with respect to the dielectric boundary. The maximum electric energy density is thus increased by a factor of

$$\frac{W_{\mathrm{e1}}}{W_{\mathrm{e0}}} = \frac{\epsilon_l |E_l|^2}{\epsilon_h |E_h|^2} \approx \frac{\epsilon_h}{\epsilon_l}, \tag{4}$$

assuming the cavity electric field is highly polarized orthogonal to the slot. Because the numerator in Eq. 1 is roughly unchanged with the introduction of the thin air slot, V_{eff} is ultimately reduced by a factor of $\sim \epsilon_h/\epsilon_l$. Recently, Seidler et. al demonstrated a silicon-air PhC cavity with an air gap to reduce the cavity mode volume by a factor of 12.1 to $V_{\mathrm{eff}} \sim 0.01\lambda^3$ [15]. The Type-1 BC is wavelength independent, which provides some tolerance to fabrication imperfections. However, applications of this 'air-mode cavity' design, which we define as a cavity with the highest electric energy density in the low index medium, have been limited because the electric field is maximized in the low-index material.

Here, we introduce a method to further reduce V_{eff} by also making use of the boundary condition on the parallel component of the electric field ('Type-2 BC'). A high-index bridge of width $b = 5$ nm is introduced across the slot of width $s = 40$ nm, as illustrated in Fig. 1(c). The parallel component of the electric field across this bridge is given by

$$E_{l||} = E_{h||}, \tag{5}$$

where $||$ represents the parallel component of the electric field. Type-2 BC forces the electric field in the bridge to be same as that in the slot. Compared to the slot cavity, the maximum electric energy density is enhanced by a factor of,

$$\frac{W_{\mathrm{e2}}}{W_{\mathrm{e1}}} = \frac{\epsilon_h |E_h|^2}{\epsilon_l |E_l|^2} \approx \frac{\epsilon_h}{\epsilon_l}. \tag{6}$$

For a vanishingly narrow bridge, the numerator of Eq. 1 is unchanged, so that V_{eff} is reduced by an additional factor of ϵ_h/ϵ_l. As opposed to the slot cavity, this modified design produces a 'dielectric-mode cavity', defined as a cavity with the highest electric energy density in the high-index dielectric. This type of cavity enables enhanced light-matter interactions with embedded emitters or the bridge material itself, that could not be covered by a slot cavity.

What happens if the bridge has finite width? For the $b = 5$ nm bridge shown in Fig. 1(c), our FDTD simulation yields $V_{\mathrm{eff}} \sim 2.5 \times 10^{-3}\lambda^3$. This mode volume is reduced by a factor of ~ 10 compared to the air-slot cavity ($2.5 \times 10^{-2}\lambda^3$) in Fig. 1(b). This factor is only slightly smaller than the analytically predicted value of $\epsilon_h/\epsilon_l = 12.1$. If b is increased to 10 nm (40 nm), as shown in Fig. 1(d) (Fig. 1(e)), the mode volume expands to $\sim 4.3(7.7) \times 10^{-3}\lambda^3$. For all of these bridge widths, V_{eff} remains below that of the original slot cavity. As the bridge width is increased, V_{eff} also increases because of a weaker effect from Type-2 BC [16]. We note that the fields outside the cavity region are nearly unchanged for these different near-field dielectric structures, i.e., they are nearly identical to Fig. 1(a). [16].

The small bridge dimensions require extremely memory-intensive and slow 3D FDTD simulations because of the requirement for nanometer-scale meshing. However, provided that the cavity modes are nearly identical in the unperturbed region in Fig. 1(a), and the EM problem is quasi-static in the deeply subwavelength scale of the cavity center, is it even necessary to perform 3D FDTD simulations to estimate the mode volume and fields? The bottom panels in Fig. 1(b)$\sim$(e) plot the electric fields obtained by 2D electrostatic simulations when an (arbitrary) potential difference ΔV is applied between the upper and lower boundaries [16]. These 2D electrostatic simulations based on finite element methods (FEM) are several orders of magnitude faster than 3D FDTD simulations. Remarkably, the $|E|$ distributions are very similar for the simple 2D FEM and the laborious 3D FDTD simulations, allowing a rapid exploration of the design space of the subwavelength dielectric structuring [16].

Combinations of Type-1 and Type-2 BCs open a wide design space. Introducing a low-index slot (i.e. exploiting the Type-1 BC) changes the cavity from a dielectric-mode cavity into an air-mode cavity. Conversely, introducing a bridge (i.e using the Type-2 BC) changes the cavity from an air-mode cavity into a dielectric-mode cavity. As a result, alternate applications of slots (Type-1 BC) and bridges (Type-2 BC) can continue reducing the mode volume. As a demonstration, Fig. 2(a) shows the cavity mode of a slot/bridge/slot cavity (called as SBS) after the addition of a $s = 1$ nm-slot (S) to the slot/bridge cavity design (SB) of Fig. 1(d), which reduces V_{eff} by ~ 7 times to $6.1 \times 10^{-4}\lambda^3$ (simulated with 3D FDTD). The

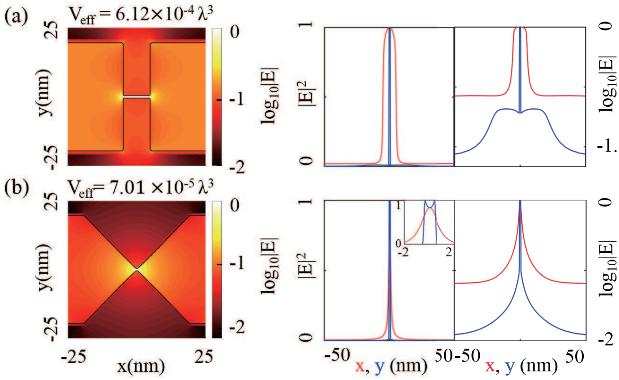

FIG. 2. Electric field distribution. (a) Slot/bridge/slot cavity (SBS). (b) Tip cavity as a limiting case of concatenation. Here, we show an air-mode cavity, and tip is smoothed out. The 2D field distribution in log scale (Left). Field distribution along the line cut in linear scale (Middle), and log scale (Right). As the same reason with Fig. 1, field at the middle of the structure is used as a maximum for $V_{\rm eff}$ calculation. Note that tip cavity has a higher field intensity at the tip than the middle in the inset.

reduction is less than ϵ_h/ϵ_l possible by the Type-1 BC because of the finite slot width, but in principle, an infinitesimal slot can achieve $\sim \epsilon_h/\epsilon_l$ reduction.

Repeated concatenations with a fixed b_i/s_i, s_{i+1}/b_i ratio, where subscript i denotes $i^{\rm th}$ step of concatenation, produces a self-similar dielectric pattern in the cavity center. Arbitrary reduction of $V_{\rm eff}$ is possible in this limit (neglecting for the moment other practical issues discussed below). In the quasi-static limit (deep subwavelength), the electric field (energy density) is only determined by the boundary conditions and (relative) permittivity distribution $\epsilon(\vec{r})$, which both have scale invariance. Thus, the expanding symmetry of the self-similar permittivity distribution implies field distribution with an expanding symmetry. This means that the electric energy density increases exponentially with the number of concatenation, resulting in vanishing $V_{\rm eff}$.

Practically, fabrication places limits on the minimum size of structures in a design, and concatenation is impossible at some point. Figure 2(b) shows the field concentration in a disconnected tip with a 45° taper angle, corresponding to a self-similar design with $b_i/s_i = 1$, $s_{i+1}/b_i = 1 - \delta$ (i=1, 2 $\ldots N$) , $\delta \to +0$, $N \to \infty$ [16]. Assuming a radius of curvature of the tip of $r = 1$ nm and a tip gap of 1 nm, we estimate a mode volume of $V_{\rm eff} = 7.0 \times 10^{-5}\lambda^3$. The panels on the right of Fig. 2 show the extreme field concentration in the horizontal (red) and vertical (blue) traces. Here we described an air-mode cavity with a disconnected tip, but the dielectric-mode cavity can also be implemented with connected tips. These designs with tip-features have the advantages in fabrication because they are easier to be fabricated than small size, 90° bridges and slots.

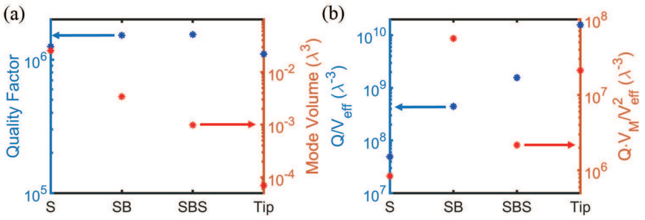

FIG. 3. (a) Quality factors and mode volumes in each case after Q optimization. S: Slot cavity ($s_1 = 40$ nm). SB: Slot/bridge cavity ($b_1 = 10$ nm). SBS: Slot/bridge/slot cavity ($s_2 = 3$ nm). Tip: Tip cavity (air-mode cavity, 1 nm gap, $r = 1$ nm) [16] (b) Two figures of merits: Q/$V_{\rm eff}$ as a general criteria and $Q \cdot V_{\rm M}/V_{\rm eff}^2$ for single-photon nonlinearities.

Electron beam lithography followed by reactive ion etching [17, 18], or focused ion beam milling [19], allow the patterning of dielectric tips with a gap below 10 nm. Alternatively, anisotropic etching of crystalline materials, such as wet etching of Si, can produce sharp tips with radius of curvature on the nanometer scale [20]. This method also has successfully demonstrated in-plane tip fabrication [21]. After, oxidation sharpening can be used further reducing the radius of curvature to sub-nanometer [22]. Lastly, the fabrication requirements are more relaxed at longer wavelengths, such as in the mid-infrared spectrum.

It is interesting to consider what happens if the tip radius r continues to be decreased. The field enhancement at a tip, with $r = 0$, is a well-studied problem in electrostatics, both for conductor [23] and dielectric [24] materials. The field at a dielectric tip, which can be expressed by a transcendental equation, diverges at the apex [24], which would result in $V_{\rm eff} = 0$, agreeing with our aforementioned proof. However, in this case, $V_{\rm eff}$ loses its physical meaning because the dipole approximation of the light-matter interaction would no longer hold.

Do the sharp features of the field concentrator still permit a high Q factor in our cavity? Robinson et al. noted that introducing a slot significantly reduced the Q factor [12]. This reduction of Q can be interpreted from perturbation theory, as the radiation loss induced by the permittivity change $\Delta\epsilon(\vec{r})$ of the PhC structure. Fortunately, it is generally possible to cancel this radiation in the far-field through additional perturbations elsewhere in the PhC structure [25]. In Fig. 3, we summarize our optimization of the Q factor of the fundamental cavity mode for successive introductions of slots and bridges (S,SB,SBS), as well as for the tip design with 1 nm gap and $r = 1$ nm. We performed these radiation loss minimizations by 3D FDTD, using particle swarm optimization for the length of the slot, the lattice constant, and the positions and radii of the holes symmetrically about the cavity center [16]. This process allowed us to maintain a high Q over 10^6 (Fig. 3(a)) across all mode concentration designs.

TABLE I. Figures of merit for application areas of photonic nanocavities.

Application	Regime	FOM
Purcell Effect	BC [a]	Q/V_{eff}
	BE [b]	$1/V_{\mathrm{eff}}$
Strong coupling	BC	$Q/\sqrt{V_{\mathrm{eff}}}$
with two level emitter	BE	$1/\sqrt{V_{\mathrm{eff}}}$
Optical bistability		Q^2/V_{Kerr} [c]
Single photon Kerr nonlinearity		$QV_{\mathrm{M}}/V_{\mathrm{eff}}^2$ [d]

[a] Bad cavity

[b] Bad emitter

[c] $V_{\mathrm{Kerr}} = \frac{(\int \epsilon |E|^2 dV)^2 \cdot \max(n_2/\epsilon)}{\int \frac{n_2 \epsilon}{3}(|E \cdot E|^2 + 2|E|^4) dV}$ [25]

[d] $V_{\mathrm{M}} = \frac{\int_M |E(\vec{r})|^4 dV}{\max(|E(\vec{r})|^4)}$

As shown in Fig. 3(b), the corresponding Q/V_{eff} values, in units of λ^{-3}, can exceed 10^{10}. It is useful to compare these Q/V_{eff} values with recently reported figures obtained by blind numerical optimizations: specifically, using an evolutionary algorithm (EA) [26], inverse design (ID) [27], and topology optimization (TO) [28]. These optimization approaches yielded Q/V_{eff} ratios of $\sim 1.0 \times 10^5$ (EA), $\sim 1.0 \times 10^6$ (ID), and $\sim 3.0 \times 10^7$(TO). The corresponding mode volumes, in units of λ^3, were 0.01 (EA), 0.007 (ID), and 0.001 (TO). These results have practical limitations. The ID and TO approaches require a continuously varying refractive index (which is difficult for commonly used materials), and all approaches produced disconnected dielectric structures (difficult for fabrication).

Remarkably, in reviewing the optimized dielectric structures from these numerical approaches, one discovers a strikingly similar feature to our designs: *two concentric tips centered at the cavity*, which are disconnected for the EA approach and joined for the ID and TO approaches [16]. Our semi-analytical analysis elucidate the origin of this feature. Optimizing this feature simplifies the design process to achieve higher Q and smaller V_{eff}, while ensuring a fully connected binary dielectric constant (that can be fabricated using standard lithography.) Also, our design suggests that even smaller mode volume is possible with a (2D-tapered) conical tip.

Table I summarizes the figures of merit (FOMs) that, in addition to Q/V_{eff}, are important for various applications, including spontaneous emission rate enhancement (Purcell effect) of quantum emitters, strong emitter-cavity coupling, optical bistability, and single-photon Kerr nonlinearities. All listed applications benefit from small V_{eff} and most benefit from high Q. Moreover, because Q factors are often practically limited (by material losses, scattering [29], or application-specific bandwidth constraints), reducing V_{eff} is particularly beneficial for many applications.

Specifically, we show here that the extreme field concentration can enable single-photon level Kerr nonlinearities at room temperature without an atomic medium or atom-like emitters, which are often difficult to fabricate and control. The Hamiltonian of a cavity with Kerr medium is expressed by [30],

$$H = [\hbar\omega - i\kappa/2 + \eta(\hat{n} - 1)]\hat{n}. \tag{7}$$

where $\kappa = \omega/Q$ is the cavity linewidth, and η is the one photon resonance frequency shift. η can be derived from perturbation theory [16],

$$\frac{\eta}{\omega} = -\frac{3\chi^{(3)}\hbar\omega}{4\epsilon_0\epsilon^2} \cdot \frac{V_{\mathrm{M}}}{V_{\mathrm{eff}}^2}. \tag{8}$$

where $V_{\mathrm{M}} = \int_M |E(\vec{r})|^4 dV / \max(|E(\vec{r})|^4)$, and the integration is over the region of nonlinear medium(M). The term $|E(\vec{r})|^4$ is due to the mode overlap ($\propto |E(\vec{r})|^2$) and the Kerr index shift ($\propto |E(\vec{r})|^2$). In our simulations, V_{M} approximates the volume of the region of highest field concentration feature (the slot, bridge, or the gap between the tip).

The condition for a single photon to shift the cavity by one resonance linewidth is [16],

$$\frac{Q \cdot V_{\mathrm{M}}}{V_{\mathrm{eff}}^2} > \frac{4\epsilon_0}{3\hbar\omega} \cdot \frac{\epsilon^2}{|\chi^{(3)}|}, \tag{9}$$

assuming the cavity radiation loss dominates over material losses (see supplemental material for a discussion on material loss [16]). Under this condition, photons in the cavity can be considered as strongly interacting particles [31]. The required $|\chi^{(3)}|/\epsilon^2$ is $\sim 1.60 \times 10^{-17}$ $\mathrm{m}^2/\mathrm{V}^2$ for the tip cavity design in Fig. 3. This type of Kerr nonlinearity is possible with organic materials that could be conveniently introduced into the air-slot of the cavity. J-aggregate (PIC) has $|\chi^{(3)}|/\epsilon^2 = 1.1 \times 10^{-15}$ $\mathrm{m}^2/\mathrm{V}^2$ at $\lambda = 575$ nm [32]; polydiacetylen (PTS) has $|\chi^{(3)}|/\epsilon^2 = -0.931 \times 10^{-17}$ $\mathrm{m}^2/\mathrm{V}^2$ at $\lambda = 1060$ nm [33]). Inorganic materials that can be deposited by atomic layer deposition (ALD) are also promising for direct introduction into the slot area; indium tin oxide (ITO), for example, has been reported to have $|\chi^{(3)}|/\epsilon^2 = 2.12 \times 10^{-17}$ $\mathrm{m}^2/\mathrm{V}^2$ at $\lambda = 1175$ nm [34].

The proposed bridge cavity requires $|\chi^{(3)}|/\epsilon^2 \sim 0.61 \times 10^{-17}$ $\mathrm{m}^2/\mathrm{V}^2$ to reach a single-photon nonlinearity. Conventional semiconductor materials such as Si (0.99×10^{-19} $\mathrm{m}^2/\mathrm{V}^2$)[35–37], GaAs ($0.97 \times 10^{-20}$ $\mathrm{m}^2/\mathrm{V}^2$, at $\lambda =$ 1.06 um) [38], Ge (0.86×10^{-20} $\mathrm{m}^2/\mathrm{V}^2$, at $\lambda = 3.17$ um) [39] do not meet this requirement, but could nevertheless produce a strong nonlinearity at extremely low powers (corresponding to a few hundreds of photons in the cavity). These parametric few-photon nonlinearities could have numerous applications in frequency conversion [40], all-optical memory, logic, and routing [41–43], neuromorphic optical computing [44, 45], and entangled photon pair production by spontaneous four-wave mixing [46–48].

In conclusion, we have introduced a recipe for ultrasmall mode volume dielectric cavity. We proposed a tip cavity structure and reviewed optimization results reported. Remarkably, the extreme field concentration enabled by these dielectric features greatly amplify nonlinear optical interactions. For realistic dielectric materials, a full cavity linewidth shift appear to be possible even for a single photon within the cavity. The ultrastrong light-matter interaction opens the door to new applications feasible even at room temperature: ultrastrong Purcell enhancement [49], single molecule sensing [7], cavity QED [50], optomechanics [51], and quantum nonlinear optics [52].

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* During writing of the manuscript, we became aware of another similar design [53].

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