New Target for Cosmic Axion Searches
Baumann, D.D.; Green, D.; Wallisch, B.

Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.117.171301

Citation for published version (APA):

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: http://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.
Supplemental Material: New Target for Cosmic Axion Searches

Daniel Baumann,1, 2 Daniel Green,3, 4 and Benjamin Wallisch1

1 DAMTP, University of Cambridge, Cambridge, CB3 0WA, United Kingdom
2 JcF, University of Amsterdam, Amsterdam, 1090 GL, Netherlands
3 Department of Physics, University of California, Berkeley, CA 94720, USA
4 Canadian Institute for Theoretical Astrophysics, Toronto, ON M5S 3H8, Canada

This document contains material supplementary to our letter New Target for Cosmic Axion Searches. In the first part, we derive the rates of Goldstone boson production that were used to obtain the results of the letter. In the second part, we discuss the (negligible) effects of decays of the massive Goldstone bosons on the derived bounds.

PRODUCTION RATES

In this part, we derive the rates of Goldstone boson production used in the main text. We consider separately the couplings to gauge fields and to matter fields.

Couplings to Gauge Fields

Above the scale of electroweak symmetry breaking (EWSB), the coupling of the Goldstone boson to the Standard Model (SM) gauge sector is

\[ \mathcal{L}_\phi = -\frac{\phi}{4\Lambda} \left( c_1 B_{\mu\nu} \tilde{B}^{\mu\nu} + c_2 W^a_{\mu\nu} \tilde{W}^{\mu\nu,a} + c_3 G^a_{\mu\nu} \tilde{G}^{\mu\nu,a} \right). \]  

(S1)

The Goldstone production associated with the couplings in (S1) was considered in [S1–S5]. In the limit of massless gauge bosons, the cross sections for some of the processes have infrared (IR) divergences, and the results depend slightly on how these divergences are regulated. In [S5], the total production rate was found to be

\[ \Gamma = \frac{T^3}{8\pi\Lambda^2} \left[ c_1^2 F_1(T) + 3c_2^2 F_2(T) + 8c_3^2 F_3(T) \right], \]  

(S2)

where the functions \( F_n(T) \) were derived numerically. We extracted \( F_n(T) \) from Fig. 1 of [S5], together with the one-loop running of the gauge couplings \( \alpha_i(T) \).

Coupling to gluons.—To isolate the effect of the coupling to gluons, we write \( c_1 = c_2 \equiv 0 \) and define \( \Lambda_g \equiv \Lambda/c_3 \). The production rate (S2) then becomes

\[ \Gamma_g(T) = \frac{F_3(T)}{\pi} \frac{T^3}{\Lambda_g^2} \equiv \gamma_g(T) \frac{T^3}{\Lambda_g^2}, \]  

(S3)

where \( \gamma_g(10^{10}\text{ GeV}) = 0.41 \). The function \( \gamma_g(T) \) is presented in the left panel of Fig. S1. The freeze-out bound on the gluon coupling then is

\[ \Lambda_g > \left( \frac{\pi^2}{90} g_{*,R} \right)^{-1/4} \sqrt{\gamma_g(T) g_{*,R}} M_{\text{pl}}, \]  

(S4)

where \( g_{*,R} \equiv g_*(T_R) \) and \( \gamma_{g,R} \equiv \gamma_g(T_R) \). The bound in (S4) is illustrated in the right panel of Fig. S1. In the main text, we used \( \Lambda_g(10^{10}\text{ GeV}) = 5.4 \times 10^{13}\text{ GeV} \).

Coupling to photons.—To isolate the coupling to the electroweak sector, we set \( c_3 = 0 \). In this case, the Lagrangian (S1) can be written as

\[ \mathcal{L}_\phi = -\frac{1}{4\Lambda} \left( c_0 B_{\mu\nu} B^{\mu\nu} + s_a W^a_{\mu\nu} \tilde{W}^{\mu\nu,a} \right), \]  

(S5)

where we have defined

\[ \Lambda = \frac{\Lambda}{\sqrt{c_1^2 + c_2^2}}, \quad c_0 = \frac{c_1}{\sqrt{c_1^2 + c_2^2}}, \quad s_a = \frac{c_2}{\sqrt{c_1^2 + c_2^2}}. \]  

(S6)

Note that \( c_0^2 + s_a^2 = 1 \), so we can use \( \Lambda \) and \( c_0 \) as the two free parameters. The production rate (S2) is then given by

\[ \Gamma = \frac{[c_0^2 F_1(T) + 3s_a^2 F_2(T)] T^3}{8\pi \Lambda^2} \equiv \gamma(T, c_0) \frac{T^3}{\Lambda^2}. \]  

(S7)

The function \( \gamma(T, c_0) \) is shown in the left panel of Fig. S2. In the main text, we employed \( \gamma(10^{10}\text{ GeV}, 1) = 0.017 \). The freeze-out bound on the coupling then is

\[ \Lambda(c_0) > \left( \frac{\pi^2}{90} g_{*,R} \right)^{-1/4} \sqrt{\gamma_R(c_0) g_{*,R}} M_{\text{pl}}, \]  

(S8)

where \( \gamma_R(c_0) \equiv \gamma(T, c_0) \). We wish to relate this bound to the couplings below the EWSB scale.

At low energies, the axion couplings to the electroweak sector become

\[ \mathcal{L}_{\text{EW}} = -\frac{1}{4} \left( \frac{\phi}{\Lambda_g} F_{\mu\nu} \tilde{F}^{\mu\nu} + \frac{\phi}{\Lambda_Z} Z_{\mu\nu} \tilde{Z}^{\mu\nu} + \frac{\phi}{\Lambda_W} W^{\mu+} \tilde{W}^{-\mu} \right), \]  

(S9)

where \( F_{\mu\nu} \), \( Z_{\mu\nu} \) and \( W^{\mu\pm} \) are the field strengths for the photon, Z and W±, respectively. Here, we have dropped additional (non-Abelian) terms proportional to \( c_2 \) which are cubic in the gauge fields. In order to match the high-energy couplings in (S5) to the low-energy couplings

\[ \text{with } \phi, \Lambda, Z, W \text{ and } F. \]  

(S10)
in \((S9)\), we define

\[
\Lambda^{-1}_\gamma = (c_w^2 c_a + s_w^2 s_a) \Lambda^{-1}, \quad (S10)
\]
\[
\Lambda^{-1}_Z = (c_w^2 s_a + s_w^2 c_a) \Lambda^{-1}, \quad (S11)
\]
\[
\Lambda^{-1}_{Z\gamma} = 2s_w c_w (s_a - c_a) \Lambda^{-1}, \quad (S12)
\]
\[
\Lambda^{-1}_W = s_a \Lambda^{-1}, \quad (S13)
\]

where \(\{c_w, s_w\} \equiv \{\cos \theta_w, \sin \theta_w\}\), with \(\theta_w \approx 30^\circ\) the Weinberg mixing angle. Using \((S10)\), we can write \((S8)\) as a bound on the photon coupling,

\[
\Lambda_\gamma(c_a) > \frac{1}{c_w^2 c_a + s_w^2 s_a} \left( \frac{\pi^2 g_{*,R}}{90} \right)^{-1/4} \sqrt{\gamma_R(c_a)} \frac{T_R}{M_{\text{Pl}}},
\]

\[
= \lambda_\gamma(T_R, c_a) \left( \frac{T_R}{10^{10} \text{GeV}} \right)^{1/2}, \quad (S14)
\]

This bound is illustrated in the right panel of Fig. S2. We see that we get the most conservative constraint by setting \(s_a = 0\), for which we have \(\lambda_\gamma(10^{10} \text{GeV}, 1) = 1.4 \times 10^{13} \text{GeV}\).

### Couplings to Charged Matter

The calculation of the Goldstone production rates associated with the couplings to the SM fermions is somewhat less developed. In this section, we will calculate the relevant rates following the procedure outlined in \([S3]\).

**Preliminaries.**—The integrated Boltzmann equation for the evolution of the number density of the Goldstone boson takes the form

\[
\dot{n}_\phi + 3H n_\phi = \Gamma(n_{\phi}^{\text{eq}} - n_\phi), \quad (S15)
\]
where \( n^a_\phi = \zeta(3)T^3/\pi^2 \) is the equilibrium density of a relativistic scalar. In order to simplify the analysis, we will replace the integration over the phase space of the final states with the center-of-mass cross section, \( \sigma_{\text{cm}} \), or the center-of-mass decay rate, \( \Gamma_{\text{cm}} \). While this approach is not perfectly accurate, it has the advantage of relating the vacuum amplitudes to the thermal production rates in terms of relatively simple integrals.

For a two-to-two process, \( 1 + 2 \rightarrow 3 + 4 \), we have

\[
\Gamma_{2\rightarrow2} \simeq \frac{1}{n^a_\phi} \int \frac{d^3p_1}{(2\pi)^3} \frac{d^3p_2}{(2\pi)^3} f_1(p_1) f_2(p_2) \frac{1}{2E_1} \frac{1}{2E_2} \left[ 1 \pm f_3 \right] \left[ 1 \pm f_4 \right] 2s \sigma_{\text{cm}}(s),
\]

(16)

where \( f_{1,2} \) are the distribution functions of the initial states and \( s \equiv (p_1 + p_2)^2 \) is the Mandelstam variable. We have included simplified Bose enhancement and Pauli blocking terms, \( \frac{1}{2} \left[ 1 \pm f_3 \right] \left[ 1 \pm f_4 \right] \), which is applicable in the center-of-mass frame where the initial and final momenta are equal.\(^1\)

\(^1\) These Pauli blocking and Bose enhancement terms were not included in [S3], as they complicate the rate calculations. We have included them to ensure that the rates computed for both the forward and backward processes give the same results.

\[\Gamma_{1\rightarrow2} \simeq \frac{1}{n^a_\phi} \int \frac{d^3p_1}{(2\pi)^3} f_1(p_1) \left[ 1 \pm f_2(p_1/2) \right] \times \left[ 1 \pm f_3(p_1/2) \right] \frac{m_3}{E_1} \Gamma_{\text{cm}},\]

(20)

where \( f_1 \) is the distribution function of the decaying particle (not necessarily \( \phi \)). We are mostly interested in the limit \( T \gg m_1 \), in which case the rate (20) reduces to

\[\Gamma_{1\rightarrow2} \simeq \frac{m_1}{T} \frac{\pi^2}{16\zeta(3)} \Gamma_{\text{cm}} \left\{ \begin{array}{ll} 1 & \text{fermion,} \\ 1 - \frac{4}{\pi^2} & \text{boson,} \end{array} \right.\]

(21)

where the dependence on the number of degrees of freedom of the decaying particle has been absorbed into \( \Gamma_{\text{cm}} \) through the sum over spins and charges. Note that, in equilibrium, the rates for decay and inverse decay are equal.

**Coupling to charged fermions.**—We consider the following coupling between a Goldstone boson and charged fermions:

\[\mathcal{L}_{\phi\psi} = \frac{\phi}{\Lambda_\psi} \left( iH \tilde{\psi}_{L,i} g_{ij} \psi_{R,j} + \text{h.c.} \right),\]

(22)

where \( g_{ij} \equiv [(\lambda_1 - \lambda_j)g_{ij} + (\lambda_i + \lambda_j)g_{ij}^\dagger], H \) is the Higgs doublet, \( \psi_{L,R} \equiv \frac{1}{2}(1 \mp \gamma^5) \psi \), and the \( SU(2)_L \) and \( SU(3)_c \) structures have been left implicit. Distinct processes dominate in the various limits of interest.

- At high energies, the Goldstone boson is produced through the following two processes (see Fig. S3): (a) \( \psi_i + \bar{\psi}_j \rightarrow H + \phi \) and (b) \( \psi_i + H \rightarrow \psi_j + \phi \). Summing over the spins and charges, we get

\[\sum |\mathcal{M}|^2(a) = 4N_\psi s g(\lambda_i, g_{ij}^\dagger),\]

(23)

\[\sum |\mathcal{M}|^2(b) = 4N_\psi s (1 - \cos \theta) g(\lambda_i, g_{ij}^\dagger),\]

(24)

where

\[g(\lambda_i, g_{ij}^\dagger) \equiv \frac{[(\lambda_1 - \lambda_j)g_{ij}^\dagger(\lambda_i + \lambda_j)^2 + (\lambda_i + \lambda_j)g_{ij}^\dagger]^2}{\Lambda_\psi^2},\]

(25)

\[\overline{\psi}_j \phi \rightarrow \psi_i H \quad \overline{\psi}_j \phi \rightarrow \psi_i \]  

(a) Fermion annihilation.  

(b) Fermion-Higgs scattering.

Figure S3. Feynman diagrams for the dominant Goldstone production via the coupling to charged fermions above the electroweak scale. For the vector and axial vector couplings, \( I \in \{V,A\} \), the "-" and "+" signs apply, respectively.
and we have combined fermion and anti-fermion scattering in the sum over charges as well as introduced

$$N_\psi \equiv \begin{cases} 1 & \psi = \text{lepton}, \\ 3 & \psi = \text{quark}. \end{cases}$$ (S26)

We also find it convenient to define $\Lambda_{ij}^I \equiv \Lambda_\psi / g_{ij}^I$, with $I \in \{ V, A \}$. Using (S17) and (S18), and treating the vector and axial-vector couplings separately, we find

$$\Gamma_{ij}^I = N_\psi \left( \frac{7}{8} \right)^2 4 \zeta(3) \frac{(\lambda_i + \lambda_j)^2}{8\pi} \frac{T^3}{(\Lambda_{ij}^I)^2} \approx 0.19 N_\psi \frac{(\lambda_i + \lambda_j)^2}{8\pi} \frac{T^3}{(\Lambda_{ij}^I)^2},$$ (S27)

where the “−” and “+” signs apply to $I = V$ and $I = A$, respectively.

- Below the scale of EWSB (which is the regime most relevant for the freeze-in constraints), the Lagrangian (S22) becomes

$$L_{\phi \psi} = i \frac{\phi}{\Lambda_\psi} \bar{\psi} \left[ (m_i - m_j) g_{ij}^V + (m_i + m_j) g_{ij}^A \right] \psi_j,$$ (S28)

where $m_i \equiv \sqrt{2} \lambda_i / v$. The Goldstone production processes associated with these couplings are shown in Fig. S4.

We first consider the diagonal part of the interaction, which takes the form $i \bar{c}_i \phi \psi_j \gamma^5 \psi_i$, with $\bar{c}_i \equiv 2 m_i g_{ij}^A / \Lambda_\psi$. Kinematical constraints require us to include at least one additional particle in order to get a non-zero amplitude. The two leading processes are (a) $\psi_i + \{ \gamma, g \} \rightarrow \psi_i + \phi$ (cf. Fig. S4a) and (b) $\psi_i + \psi_i \rightarrow \psi_i + \phi + \{ \gamma, g \}$ (cf. Fig. S4b), where $\{ \gamma, g \}$ is either a photon or gluon depending on whether the fermion is a lepton or quark, respectively. Summing over spins and charges, we obtain

$$\sum |M|_{(a)}^2 = 16\pi A_\psi |\bar{c}_{ii}|^2 \frac{s^2}{(s - t)(m_i^2 - u)},$$ (S29)

$$\sum |M|_{(b)}^2 = 16\pi A_\psi |\bar{c}_{ii}|^2 \frac{t^2}{(s - m_i^2)(m_i^2 - u)},$$ (S30)

where $s, t, u$ are the Mandelstam variables and

$$A_\psi \equiv \begin{cases} \alpha & \psi = \text{lepton}, \\ 4\alpha_s & \psi = \text{quark}. \end{cases}$$ (S31)

In the massless limit, the cross section has IR divergences in the $t$- and $u$-channels from the exchange of a massless fermion. The precise production rate therefore depends on the treatment of the soft modes. Regulating the IR divergence with the fermion mass and taking the limit $s \gg m_i^2$, we find

$$\sigma_{cm}(s) \approx \frac{1}{s} A_\psi |\bar{c}_{ii}|^2 \left[ 3 \log \frac{s}{m_i^2} - \frac{3}{2} \right].$$ (S32)

At high temperatures, the fermion mass is controlled by the thermal mass $m_i^2 \rightarrow m_T^2 = \frac{1}{2} \pi A_\psi T^2$ and the production rate becomes

$$\tilde{\Gamma}_{ii} = \frac{3\pi^3}{64\zeta(3)} A_\psi |\bar{c}_{ii}|^2 \frac{T}{8\pi} \left[ \log \frac{2}{\pi A_\psi} + 2 \log 2 - \frac{3}{2} \right].$$ (S33)

This formula is expected to break down at $T \lesssim m_i$, but will be sufficient at the level of approximation being used in this paper. A proper treatment of freeze-in at $T \sim m_i$ should go beyond $\Gamma = H$ and fully solve the Boltzmann equations. However, this level of accuracy isn’t needed for estimating the constraint on the coupling $\bar{c}_{ii}$.

The result (S33) will be of limited utility for the coupling to quarks. This is because, for $T \lesssim 30$ GeV, the QCD coupling becomes large and our perturbative calculation becomes unreliable. In fact, we see that the production rate (S33) becomes negative in this regime. While the top quark is sufficiently heavy to be still at weak coupling, its mass is close to the electroweak phase transition and, therefore, the assumption $s \gg m_T^2$ is not applicable. For these reasons, we will not derive bounds on the quark couplings from these production rates.

When the coupling of $\phi$ is off-diagonal in the mass basis, the dominant process at low energies is the decay $\psi_i \rightarrow \psi_j + \phi$, cf. Fig. S4c. Since the mass splittings of the

\[ \text{Figure S4. Feynman diagrams for the dominant Goldstone production via the coupling to charged fermions below the electroweak scale. For quarks, the coupling to photons is replaced by that to gluons. In addition to the displayed s- and t-channel diagrams for the Compton-like process and fermion annihilation, there are u-channel diagrams which are not shown.} \]

\[ \text{At high temperatures, the fermion mass is controlled by} \]

\[ \text{the thermal mass} \quad m_i^2 \rightarrow m_T^2 = \frac{1}{2} \pi A_\psi T^2 \]

\[ \text{and the production rate becomes} \]

\[ \tilde{\Gamma}_{ii} = \frac{3\pi^3}{64\zeta(3)} A_\psi |\bar{c}_{ii}|^2 \frac{T}{8\pi} \left[ \log \frac{2}{\pi A_\psi} + 2 \log 2 - \frac{3}{2} \right]. \]

\[ \text{This formula is expected to break down at} \quad T \lesssim m_i, \quad \text{but will be sufficient at the level of approximation being used in this paper. A proper treatment of freeze-in at} \quad T \sim m_i \quad \text{should go beyond} \quad \Gamma = H \quad \text{and fully solve the Boltzmann equations. However, this level of accuracy isn’t needed for estimating the constraint on the coupling} \quad \bar{c}_{ii}. \]

\[ \text{The result} \quad (S33) \quad \text{will be of limited utility for the coupling to quarks. This is because, for} \quad T \lesssim 30 \text{ GeV, the QCD coupling becomes large and our perturbative calculation becomes unreliable. In fact, we see that the production rate} \quad (S33) \quad \text{becomes negative in this regime. While the top quark is sufficiently heavy to be still at weak coupling, its mass is close to the electroweak phase transition and, therefore, the assumption} \quad s \gg m_T^2 \quad \text{is not applicable. For these reasons, we will not derive bounds on the quark couplings from these production rates.} \]

\[ \text{When the coupling of} \quad \phi \quad \text{is off-diagonal in the mass basis, the dominant process at low energies is the decay} \quad \psi_i \rightarrow \psi_j + \phi, \text{cf. Fig. S4c. Since the mass splittings of the} \]

\[ \text{effects are computable using the techniques of} \quad [S5], \quad \text{but this is beyond the scope of the present work.} \]
SM fermions are large and $m_\phi \ll m_\psi$, the center-of-mass decay rate is well approximated by
$$\Gamma_{\text{cm}} = \frac{N_\psi m_\phi^3}{8\pi \Lambda_{ij}^2},$$
where $\Lambda_{ij} \equiv [\langle g_{ij}^\Lambda \rangle^2 + \langle g_{ij}^V \rangle^2]^{-1/2} \Lambda_\psi$. Using (S21), we get
$$\Gamma_{ij} = \frac{(\pi^2 - 4)}{16\zeta(3)} \frac{N_\psi m_\phi^4}{8\pi \frac{m_\phi^2}{T} \Lambda_{ij}^2} \simeq 0.31N_\psi \frac{|\tilde{\epsilon}_{ij}|^2 m_\phi^2}{8\pi T},$$
with $|\tilde{\epsilon}_{ij}| \approx m_\phi/\Lambda_{ij}$. In addition to this decay, we also have production with a photon or gluon, given by (S33) with $\tilde{\epsilon}_{ii} \rightarrow \tilde{\epsilon}_{ij}$. We will neglect this contribution as it is suppressed by a factor of $\alpha$ or $\alpha_s$ for $T \sim m_\psi$.

**COMMENTS ON DECAYS**

We have treated each of the operators which couple the pNGBs to the SM independently throughout. For computing the production rates, this is justified since the amplitudes for the different processes that we consider do not interfere and the couplings therefore add in quadrature. One may still ask, however, if the interplay between several operators can affect the cosmological evolution after the production. In particular, one might worry that some operators would allow for the decay of the pNGBs and that this might evade the limits on $N_{\text{eff}}$. In this part, we will address this concern. We are assuming that $m_\phi < 1$ MeV, so that the only kinematically allowed decays are to photons and neutrinos.

**Decay to Photons**

The coupling $\phi F \bar{F}$ can mediate the decay of axions to photons. However, for the range of parameters of interest, these decays occur after recombination and, hence, do not affect the CMB. To see this, we consider the decay rate for $m_\phi \gtrsim T$ [S6],
$$\Gamma_{D,\gamma} = \frac{1}{64\pi} \frac{m_\phi^3}{\Lambda_\gamma^2}.$$  
(S36)

The decay time is $T_D = \Gamma_{D,\gamma}^{-1}$ and the temperature at decay is determined by $H (T_D) \approx \Gamma_{D,\gamma}^{-1}$. We will not consider the regime $m_\phi < T_D$ as it does not arise in the range of parameters of interest. Assuming that the universe is matter dominated at the time of the decay, we get
$$\frac{T_D}{T_{\text{rec}}} \approx 9.5 \times 10^{-10} \left( \frac{\Lambda_\gamma}{10^{10} \text{GeV}} \right)^{-4/3} \left( \frac{m_\phi}{T_{\text{rec}}} \right)^2.$$  
(S37)

Recalling the constraint from stellar cooling, $\Lambda_\gamma > 1.3 \times 10^{10}$ GeV [S7], we therefore infer that $T_D < 7.1 \times 10^{-10} T_{\text{rec}} (m_\phi/T_{\text{rec}})^2$, so that the axions are stable on the time-scale of recombination as long as $m_\phi \lesssim 10$ keV. CMB-S4 will probe this regime through sensitivity to $N_{\text{eff}}$ for $m_\phi < T_{\text{rec}}$ and through sensitivity to warm dark matter for larger masses. Warm dark matter is already highly constrained by cosmology, with values of $m_\phi$ above 1 eV typically ruled out by the CMB. For comparison, a stable particle with $m_\phi \geq 100$ eV produces $\Omega_m > 1$ and is therefore excluded by constraints on the dark matter abundance. For $m_\phi > 10$ keV, the decay to photons does affect the phenomenology and must be considered explicitly. Nevertheless, in the regime of interest, $10$ keV < $m_\phi < 1$ MeV (where the axion decays between neutrino decoupling and recombination), the pNGBs are non-relativistic and, therefore, carry a large energy density, $\rho_\phi \approx m_\phi n_\phi$. As a result, this region is highly constrained by current cosmological observations [S8, S9].

**Decay to Neutrinos**

Depending on the mass of the pNGB, the decay to neutrinos leads to the following three scenarios:

- For $m_\phi < T_{\text{rec}}$, the strong interactions between the pNGBs and the neutrinos imply that the neutrinos are no longer free-streaming particles [S10-S12], which is ruled out by recent CMB observations [S13].
- For $T_D > m_\phi > T_{\text{rec}}$, the pNGBs are brought into equilibrium with the neutrinos at $T \sim T_D$ and then become Boltzmann suppressed for $T \lesssim m_\phi$. This process leads to a contribution to $N_{\text{eff}}$, even if the pNGBs have negligible energy density to begin with. To estimate the size of the effect, we first note that the freeze-in at $T_D$ conserves the total energy density in neutrinos and pNGBs,
$$\langle g_\nu + g_\phi \rangle (a_1 T_1)^4 = g_\nu (a_0 T_0)^4,$$  
(S38)

where $T_0$ and $T_1$ are the initial and final temperatures during the equilibration, and $g_\nu$ and $g_\phi = 1$ are the effective numbers of degrees of freedom in $\nu$ and $\phi$, respectively. When the temperature drops below the mass of the pNGBs, their energy density is converted to neutrinos. This process conserves the comoving entropy density,
$$\langle g_\nu + g_\phi \rangle (a_1 T_1)^3 = g_\nu (a_2 T_2)^3,$$  
(S39)

where $T_2 \lesssim m_\phi$ is some temperature after the pNGB population has decayed. The final energy density of the neutrinos becomes
$$a_4^4 \rho_{\nu,2} = \left( \frac{g_\nu + g_\phi}{g_\nu} \right)^{1/3} a_0^4 \rho_{\nu,0},$$  
(S40)

where $\rho_{\nu,i} \equiv \rho_\nu (a_i)$. Using the definition of $N_{\text{eff}}$ in (2) of
the main text and \( a^4 \rho_\gamma = \text{const.} \), we find
\[
N_{\text{eff}} = \left( \frac{g_\nu + g_\phi}{g_\nu} \right)^{1/3} N_{\text{eff},0}.
\] (S41)

Considering the coupling to a single neutrino flavor (rather than all three), i.e. \( N_{\text{eff},0} \simeq 1 \) and \( g_\nu = 7/4 \), we then get
\[
\Delta N_{\text{eff}} = \left(1 + \frac{4}{7}\right)^{1/3} - 1 \simeq 0.16,
\] (S42)

where \( \Delta N_{\text{eff}} \equiv N_{\text{eff}} - N_{\text{eff},0} \). Coupling to more than one neutrino flavor and including a non-zero initial temperature for the pNGBs would increase this number slightly, so that we will use \( \Delta N_{\text{eff}} \geq 0.16 \).

- The production of pNGBs through the freeze-in process is avoided if \( m_\phi > T_D > T_{\text{rec}} \), in which case the pNGBs decay to neutrinos out of equilibrium. To a good approximation, this decay conserves the energy density, which is therefore simply transferred from \( \phi \) to \( \nu \) at the time of the decay. The contribution to \( \Delta N_{\text{eff}} \) is enhanced by the amount of time that \( \phi \) is non-relativistic before its decay, which may be a large effect for \( m_\phi \gg 1 \text{ eV} \).

In summary, operators that allow the Goldstone bosons to decay do not substantially alter the predictions presented in the main text. On the one hand, decays to photons cannot occur early enough to impact the CMB. On the other hand, decays to neutrinos typically increase the contributions to \( \Delta N_{\text{eff}} \) and would therefore strengthen our bounds.

---