New Members of SHG Active Dugganite Family, $A_3BC_3D_2O_{14}$ ($A =$ Ba, Pb; $B =$ Te, Sb; $C =$ Al, Ga, Fe, Zn; $D =$ Si, Ge, P, V): Synthesis, Structure and Materials Properties

Anupam Bhim,† Weiguo Zhang,‡ P. Shiv Halasyamani,*‡ Jagannatha Gopalakrishnan*,† and Srinivasan Natarajan*,†

†Solid State and Structural Chemistry Unit, Indian Institute of Science, Bangalore–560012, India
‡Department of Chemistry, University of Houston, 112 Fleming Building, Houston, Texas 77204–5003, United States

Electronic Supporting Information

Table of contents

1. Table S1 Details of the synthesis conditions for the compounds……………………………………………..S3
2. Figure S1 SEM image and corresponding EDX spectrum of the compounds………………………………S4-S6
3. Figure S2-S11 Rietveld refinements of the compounds………………………………………………………S6-S11
4. Table S2-S11 Crystallographic data of the compounds……………………………………………………..S11-S16
5. Figure S12 Crystal structure (ball-and-stick representation) of $Ba_3TeGa_2ZnSi_2O_{14}$……………………...S16
6. Figure S13 Phase-matching curves for moderate phase-matchable SHG active compounds.................S17
7. Figure S14 Dielectric constant and dielectric loss versus frequency plots for all ………………………S17
8. Figure S15. Comparison of UV/Vis and NIR reflectance (%) of dugganite compounds with TiO$_2$…S18
9. Figure S16. PXRD patterns of different ($Ba/Pb$)$_3$Te($Ga/Fe$)$_2$ZnM$^{III}$Si$_2$O$_{14}$ ($M^{III}=$Co, Ni, and Cu) ...S18
10. Figure S17. Colors of ($Ba/Pb$)$_3$Te($Ga/Fe$)$_2$ZnM$^{III}$Si$_2$O$_{14}$ ($M^{III}=$Co, Ni, and Cu) under daylight......S19
11. Figure S18. Optical absorption (UV/Vis) spectra of ($Ba/Pb$)$_3$TeFe$^2$CoSi$_2$O$_{14}$..............................S19
12. Figure S19. CIE chromaticity diagram for ($Ba/Pb$)$_3$Te($Ga/Fe$)$_2$ZnM$^{III}$Si$_2$O$_{14}$ ($M^{III}=$Co, Ni, and Cu) S20
13. Table S12. Color coordinates for ($Ba/Pb$)$_3$Te($Ga/Fe$)$_2$ZnM$^{III}$Si$_2$O$_{14}$ ($M^{III}=$Co, Ni, and Cu) ………S20
14. Figure S20. Partial density of states of the compounds…………………………………………………………S21

S1
Table S1. Details of the synthesis conditions for the single phase Dugganite compounds

<table>
<thead>
<tr>
<th>Compound</th>
<th>Temperature (°C)/Time (h)</th>
<th>Color</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba₃TeGa₂ZnSi₂O₁₄</td>
<td>1075/24</td>
<td>white</td>
</tr>
<tr>
<td>Ba₃TeFe₂ZnSi₂O₁₄</td>
<td>1060/24</td>
<td>beige</td>
</tr>
<tr>
<td>Pb₃TeGa₂ZnSi₂O₁₄</td>
<td>720/24</td>
<td>white</td>
</tr>
<tr>
<td>Pb₃TeFe₂ZnSi₂O₁₄</td>
<td>710/24</td>
<td>beige</td>
</tr>
<tr>
<td>Pb₃TeAl₂ZnSi₂O₁₄</td>
<td>730/24</td>
<td>white</td>
</tr>
<tr>
<td>Pb₃TeGa₂ZnGe₂O₁₄</td>
<td>710/24</td>
<td>white</td>
</tr>
<tr>
<td>Pb₃TeFe₂ZnGe₂O₁₄</td>
<td>700/24</td>
<td>yellow</td>
</tr>
<tr>
<td>Pb₃SbGaZn₂P₂O₁₄</td>
<td>800/24</td>
<td>white</td>
</tr>
<tr>
<td>Pb₃SbFeZn₂P₂O₁₄</td>
<td>790/24</td>
<td>yellowish</td>
</tr>
<tr>
<td>PbBa₂TeZn₃V₂O₁₄</td>
<td>870/48</td>
<td>white</td>
</tr>
<tr>
<td>Pb₂BaTeZn₃V₂O₁₄</td>
<td>780/48</td>
<td>white</td>
</tr>
</tbody>
</table>
Figure S1. SEM image and corresponding EDX spectrum of (a) Ba$_3$TeGa$_2$ZnSi$_2$O$_{14}$ (b) Ba$_3$TeFe$_2$ZnSi$_2$O$_{14}$ (c) Pb$_3$TeGa$_2$ZnSi$_2$O$_{14}$ (d) Pb$_3$TeFe$_2$ZnSi$_2$O$_{14}$ (e) Pb$_3$TeAl$_2$ZnSi$_2$O$_{14}$ (f) Pb$_3$SbGaZn$_3$P$_2$O$_{14}$ (g) Pb$_3$SbFeZn$_2$P$_2$O$_{14}$ (h) Pb$_3$TeGa$_2$ZnGe$_2$O$_{14}$ (i) Pb$_3$TeFe$_2$ZnGe$_2$O$_{14}$ (j) PbBa$_2$TeZn$_3$V$_2$O$_{14}$

Figure S2. Rietveld refinement of Ba$_3$TeFe$_2$ZnSi$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (|) indicate Bragg reflections.
Figure S3. Rietveld refinement of Pb$_3$TeGa$_2$ZnSi$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (I) indicate Bragg reflections.

Figure S4. Rietveld refinement of Pb$_3$TeFe$_2$ZnSi$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (I) indicate Bragg reflections.
Figure S5. Rietveld refinement of Pb$_3$TeAl$_2$ZnSi$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars ($) indicate Bragg reflections.

Figure S6. Rietveld refinement of Pb$_3$TeFe$_2$ZnGe$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars ($) indicate Bragg reflections.
Figure S7. Rietveld refinement of \( \text{Pb}_3\text{TeGa}_2\text{ZnGe}_2\text{O}_{14} \) from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (\( | \)) indicate Bragg reflections.

Figure S8. Rietveld refinement of \( \text{Pb}_3\text{SbGaZn}_2\text{P}_2\text{O}_{14} \) from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (\( | \)) indicate Bragg reflections.
Figure S9. Rietveld refinement of Pb$_3$SbFeZn$_2$P$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (|) indicate Bragg reflections.

Figure S10. Rietveld refinement of Pb$_2$BaTeZn$_3$V$_2$O$_{14}$ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (|) indicate Bragg reflections.
Figure S11. Rietveld refinement of PbBa₂TeZn₇V₂O₁₄ from the PXRD data. The observed (O), calculated (red line), and difference (bottom blue line) profiles are shown. The vertical bars (|) indicate Bragg reflections.

Table S2. Crystallographic data for Ba₃TeFe₂ZnSi₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U_iso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba</td>
<td>3e</td>
<td>0.442(3)</td>
<td>0.0</td>
<td>0.0</td>
<td>0.010(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.013(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Fe/Zn</td>
<td>3f</td>
<td>0.756(4)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.003(2)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Si</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.472(3)</td>
<td>0.003(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.763(2)</td>
<td>0.009(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.492(3)</td>
<td>0.300(2)</td>
<td>0.637(2)</td>
<td>0.002(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.220(9)</td>
<td>0.102(2)</td>
<td>0.228(4)</td>
<td>0.032(3)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group $P321$: $a = b = 8.520(1)$ Å, $c = 5.234 (1)$ Å, $α = β = 90°$, $γ = 120°$;

Reliability Factors: $R_p = 2.23\%$, $R_{wp} = 3.24\%$, $χ^2 = 4.52$;

Bond Lengths (Å):

Fe/Zn–O = 1.931(1), Te–O = 2.02(1), Si–O = 1.494(2),
Ba–O = 2.825 (average)
Table S3. Crystallographic data for Pb₃TeGa₂ZnSi₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U_iso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.426(3)</td>
<td>0.426(3)</td>
<td>1.0</td>
<td>0.033(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.021(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>Ga/Zn</td>
<td>3f</td>
<td>0.254(3)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.018(2)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Si</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.529(2)</td>
<td>0.009(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.220(5)</td>
<td>0.009(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.221(8)</td>
<td>0.123(3)</td>
<td>0.767(1)</td>
<td>0.021(5)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.189(4)</td>
<td>0.497(2)</td>
<td>0.652(6)</td>
<td>0.038(1)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P₃21: a = b = 8.405(1) Å, c = 5.090 (1) Å, α= β = 90°, γ =120°;
Reliability Factors: Rp = 3.51%, Rwp = 4.57%, χ² = 2.38;
Bond Lengths (Å):
Ga/Zn–O = 1.901(1), Te–O = 2.002(1), Si–O = 1.497(2),
Pb–O = 2.741 (average)

Table S4. Crystallographic data for Pb₃TeFe₂ZnSi₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U_iso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.425(4)</td>
<td>0.425(4)</td>
<td>1.0</td>
<td>0.032(5)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.025(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Fe/Zn</td>
<td>3f</td>
<td>0.252(1)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.021(6)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Si</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.508(4)</td>
<td>0.009(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.210(3)</td>
<td>0.006(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.220(9)</td>
<td>0.120(4)</td>
<td>0.763(10)</td>
<td>0.038(7)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.185(5)</td>
<td>0.499(1)</td>
<td>0.640(2)</td>
<td>0.063(4)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P₃21: a = b = 8.419(1) Å, c = 5.146 (1) Å, α= β = 90°, γ =120°;
Reliability Factors: Rp = 2.11%, Rwp = 3.18%, χ² = 6.25;
Bond Lengths (Å):
Fe/Zn–O = 1.909(1), Te–O = 2.018(1), Si–O = 1.507(2),
Pb–O = 2.755 (average)
Table S5. Crystallographic data for Pb₃TeAl₂ZnSi₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>Uiso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.425(7)</td>
<td>0.425(7)</td>
<td>1.0</td>
<td>0.032(6)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.020(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>Al/Zn</td>
<td>3f</td>
<td>0.259(5)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.013(7)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Si</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.529(1)</td>
<td>0.012(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.232(3)</td>
<td>0.011(6)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.221(4)</td>
<td>0.120(4)</td>
<td>0.768(2)</td>
<td>0.001(5)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.192(3)</td>
<td>0.475(8)</td>
<td>0.632(9)</td>
<td>0.068(3)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P321: \(a = b = 8.365(1) \text{ Å}, c = 5.93(1) \text{ Å}, \alpha = \beta = 90^\circ, \gamma = 120^\circ;\)

Reliability Factors: \(R_p = 3.17\%, \ R_{wp} = 3.83\%, \chi^2 = 6.45;\)

Bond Lengths (Å):

Al/Zn–O = 1.887(1), Te–O = 1.991(2), Si–O = 1.52(2),

Pb–O = 2.749 (average)

Table S6. Crystallographic data for Pb₃TeFe₂ZnGe₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>Uiso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.423(2)</td>
<td>0.423(2)</td>
<td>1.0</td>
<td>0.037(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.026(9)</td>
<td>1.0</td>
</tr>
<tr>
<td>Fe/Zn</td>
<td>3f</td>
<td>0.242(1)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.027(4)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Ge</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.514(2)</td>
<td>0.024(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.195(5)</td>
<td>0.022(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.214(2)</td>
<td>0.120(9)</td>
<td>0.783(7)</td>
<td>0.016(8)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.172(8)</td>
<td>0.497(12)</td>
<td>0.650(3)</td>
<td>0.066(1)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P321: \(a = b = 8.502(1) \text{ Å}, c = 5.177(1) \text{ Å}, \alpha = \beta = 90^\circ, \gamma = 120^\circ;\)

Reliability Factors: \(R_p = 2.31\%, \ R_{wp} = 2.42\%, \chi^2 = 4.12;\)

Bond Lengths (Å):

Fe/Zn–O = 1.921(1), Te–O = 1.981(2), Ge–O = 1.56(1),

Pb–O = 2.742 (average)
Table S7. Crystallographic data for Pb$_3$TeGa$_2$ZnGe$_2$O$_{14}$.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U$_{iso}$(Å$^2$)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.422</td>
<td>0.422</td>
<td>1.0</td>
<td>0.037(5)</td>
<td>1.0</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.022(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>Ga/Zn</td>
<td>3f</td>
<td>0.250</td>
<td>0.0</td>
<td>0.5</td>
<td>0.017(1)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Ge</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.476(8)</td>
<td>0.021(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.208(3)</td>
<td>0.091(11)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.215</td>
<td>0.121</td>
<td>0.771(4)</td>
<td>0.019(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.175</td>
<td>0.459</td>
<td>0.668(7)</td>
<td>0.041(1)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group $P\overline{3}21$: $a = b = 8.523(1)$ Å, $c = 5.116(1)$ Å, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$;
Reliability Factors: $R_p = 3.26\%$, $R_{wp} = 4.64\%$, $\chi^2 = 4.782$;
Bond Lengths (Å):
Ga/Zn–O = 1.919(1), Te–O = 1.978(1), Ge–O = 1.57(2),
Pb–O = 2.744(average)

Table S8. Crystallographic data for Pb$_3$SbGaZn$_2$P$_2$O$_{14}$.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U$_{iso}$(Å$^2$)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.406</td>
<td>0.406</td>
<td>1.0</td>
<td>0.022(8)</td>
<td>1.0</td>
</tr>
<tr>
<td>Sb</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.015(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Ga/Zn</td>
<td>3f</td>
<td>0.247</td>
<td>0.0</td>
<td>0.5</td>
<td>0.021(9)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>P</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.544(2)</td>
<td>0.017(6)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.234(7)</td>
<td>0.034(3)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.206</td>
<td>0.121</td>
<td>0.756(3)</td>
<td>0.067(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.199</td>
<td>0.485</td>
<td>0.652(2)</td>
<td>0.032(3)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group $P\overline{3}21$: $a = b = 8.379(2)$ Å, $c = 5.182(1)$ Å, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$;
Reliability Factors: $R_p = 2.61\%$, $R_{wp} = 3.62\%$, $\chi^2 = 6.197$;
Bond Lengths (Å):
Ga/Zn–O = 1.911(3), Sb–O = 1.969(1), P–O = 1.505(1),
Pb–O = 2.741(average)
Table S9. Crystallographic data for Pb₃SbFeZn₂P₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U_iso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>3e</td>
<td>0.408(7)</td>
<td>0.408(7)</td>
<td>1.0</td>
<td>0.029(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>Sb</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.026(3)</td>
<td>1.0</td>
</tr>
<tr>
<td>Fe/Zn</td>
<td>3f</td>
<td>0.247(1)</td>
<td>0.0</td>
<td>0.5</td>
<td>0.027(2)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>P</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.545(4)</td>
<td>0.008(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.333</td>
<td>0.667</td>
<td>0.232(3)</td>
<td>0.057(7)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.207(5)</td>
<td>0.121(8)</td>
<td>0.752(6)</td>
<td>0.055(9)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.198(12)</td>
<td>0.488(7)</td>
<td>0.660(4)</td>
<td>0.058(2)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P321: a = b = 8.366(2) Å, c = 5.202(1) Å, α = β = 90°, γ = 120°;
Reliability Factors: R_p = 2.24%, R_wp = 3.02%, χ² = 5.423;
Bond Lengths (Å):
Fe/Zn–O = 1.916(2), Sb–O = 1.984(1), P–O = 1.512(2),
Pb–O = 2.737(average)

Table S10. Crystallographic data for Pb₂BaTeZn₃V₂O₁₄.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>U_iso(Å²)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb/Ba</td>
<td>3e</td>
<td>0.405(4)</td>
<td>0.405(4)</td>
<td>1.0</td>
<td>0.040(7)</td>
<td>0.666/0.334</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.032(8)</td>
<td>1.0</td>
</tr>
<tr>
<td>Zn</td>
<td>3f</td>
<td>0.0</td>
<td>0.237(8)</td>
<td>0.5</td>
<td>0.027(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>V</td>
<td>2d</td>
<td>0.667</td>
<td>0.333</td>
<td>0.511(9)</td>
<td>0.020(6)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.667</td>
<td>0.333</td>
<td>0.209(4)</td>
<td>0.024(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.127(4)</td>
<td>0.210(8)</td>
<td>0.777(5)</td>
<td>0.107(1)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.461(7)</td>
<td>0.191(2)</td>
<td>0.670(6)</td>
<td>0.035(3)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group P321: a = b = 8.634(2) Å, c = 5.224(1) Å, α = β = 90°, γ = 120°;
Reliability Factors: R_p = 2.50%, R_wp = 3.38%, χ² = 5.709;
Bond Lengths (Å):
Zn–O = 1.974(2), Te–O = 1.968(1), V–O = 1.702(1),
Pb/Ba–O = 2.747(average)
Table S11. Crystallographic data for PbBa$_2$TeZn$_3$V$_2$O$_{14}$.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Site</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>$U_{iso}$(Å$^2$)</th>
<th>Occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb/Ba</td>
<td>3e</td>
<td>0.394(8)</td>
<td>0.394(8)</td>
<td>1.0</td>
<td>0.035(3)</td>
<td>0.334/0.666</td>
</tr>
<tr>
<td>Te</td>
<td>1a</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
<td>0.023(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>Zn</td>
<td>3f</td>
<td>0.0</td>
<td>0.230(4)</td>
<td>0.5</td>
<td>0.023(8)</td>
<td>1.0</td>
</tr>
<tr>
<td>V</td>
<td>2d</td>
<td>0.667</td>
<td>0.333</td>
<td>0.530(11)</td>
<td>0.024(7)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>2d</td>
<td>0.667</td>
<td>0.333</td>
<td>0.222(7)</td>
<td>0.083(4)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>6g</td>
<td>0.124(2)</td>
<td>0.212(6)</td>
<td>0.780(3)</td>
<td>0.067(9)</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>6g</td>
<td>0.468(6)</td>
<td>0.189(5)</td>
<td>0.668(4)</td>
<td>0.040(1)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space group $P321$: $a = b = 8.664(2)$ Å, $c = 5.262(1)$ Å, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$;
Reliability Factors: $R_p = 2.28\%$, $R_{wp} = 3.68\%$, $\chi^2 = 3.123$;
Bond Lengths (Å):
Zn–O = 1.981(2), Te–O = 1.975(1), V–O = 1.691(2),
Pb/Ba–O = 2.769(average)

Figure S12. Crystal structure of Ba$_3$TeGa$_2$ZnSi$_2$O$_{14}$
Figure S13. Phase-matching data for moderate phase-matchable SHG active new Dugganite compounds. Inset showing non-phase-matchable SHG active compound, Pb$_3$SbGaZn$_2$P$_2$O$_{14}$. The solid curves are drawn to guide the eye and are not a fit to the data.

Figure S14. (a) The dielectric constant and (b) dielectric loss versus frequency plots for all Dugganites at room temperature.
Figure S15. Comparison of UV/Vis and NIR reflectance (%) of dugganite compounds with TiO$_2$.

Figure S16. PXRD patterns of different (Ba/Pb)$_3$Te(Ga/Fe)$_2$ZnM$^{II}$Si$_2$O$_{14}$ (M$^{II}$=Co, Ni, and Cu)
Figure S17. Colors of (Ba/Pb)$_3$TeGa$_2$ZnM$^{II}$Si$_2$O$_{14}$ (M$^{II}$=Co, Ni, and Cu) under daylight.

Figure S18. Optical absorption (UV/Vis) spectra of (Ba/Pb)$_3$TeFe$_2$CoSi$_2$O$_{14}$. 
Figure S19. CIE chromaticity diagram for (Ba/Pb)$_3$Te(Ga/Fe)$_2$ZnM$^{II}$Si$_2$O$_{14}$ (M$^{II}$=Co, Ni, and Cu) members.

Table S12. Color coordinates for (Ba/Pb)$_3$Te(Ga/Fe)$_2$ZnM$^{II}$Si$_2$O$_{14}$ (M$^{II}$=Co, Ni, and Cu) members.

<table>
<thead>
<tr>
<th>Compound</th>
<th>x</th>
<th>y</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba$_3$TeGa$_2$CoSi$<em>2$O$</em>{14}$</td>
<td>0.16</td>
<td>0.06</td>
</tr>
<tr>
<td>Ba$_3$TeGa$_2$NiSi$<em>2$O$</em>{14}$</td>
<td>0.33</td>
<td>0.14</td>
</tr>
<tr>
<td>Ba$_3$TeGa$_2$CuSi$<em>2$O$</em>{14}$</td>
<td>0.36</td>
<td>0.39</td>
</tr>
<tr>
<td>Ba$_3$TeFe$_2$ZnSi$<em>2$O$</em>{14}$</td>
<td>0.45</td>
<td>0.44</td>
</tr>
<tr>
<td>Ba$_3$TeFe$_2$CoSi$<em>2$O$</em>{14}$</td>
<td>0.23</td>
<td>0.52</td>
</tr>
<tr>
<td>Pb$_3$TeGa$_2$CoSi$<em>2$O$</em>{14}$</td>
<td>0.26</td>
<td>0.24</td>
</tr>
<tr>
<td>Pb$_3$TeGa$_2$NiSi$<em>2$O$</em>{14}$</td>
<td>0.39</td>
<td>0.16</td>
</tr>
<tr>
<td>Pb$_3$TeGa$_2$CuSi$<em>2$O$</em>{14}$</td>
<td>0.36</td>
<td>0.57</td>
</tr>
<tr>
<td>Pb$_3$TeFe$_2$ZnSi$<em>2$O$</em>{14}$</td>
<td>0.49</td>
<td>0.41</td>
</tr>
<tr>
<td>Pb$_3$TeFe$_2$CoSi$<em>2$O$</em>{14}$</td>
<td>0.12</td>
<td>0.59</td>
</tr>
</tbody>
</table>
Figure S20. Partial density of states for (a) Pb$_3$TeAl$_2$ZnSi$_2$O$_{14}$, (b) Pb$_3$TeGa$_2$ZnGe$_2$O$_{14}$, (c) Ba$_3$TeFe$_2$ZnSi$_2$O$_{14}$ (d) Pb$_3$TeFe$_2$ZnSi$_2$O$_{14}$ (e) Pb$_3$TeFe$_2$ZnGe$_2$O$_{14}$ (f) Pb$_3$SbFeZn$_2$P$_2$O$_{14}$ (g) PbBa$_2$TeZn$_3$V$_2$O$_{14}$. The Fermi level is set to 0 eV (broken black line).