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Supporting Information

$IrSi_3As_3$: First Transition Metal Arsenide Non-Linear Optical Material

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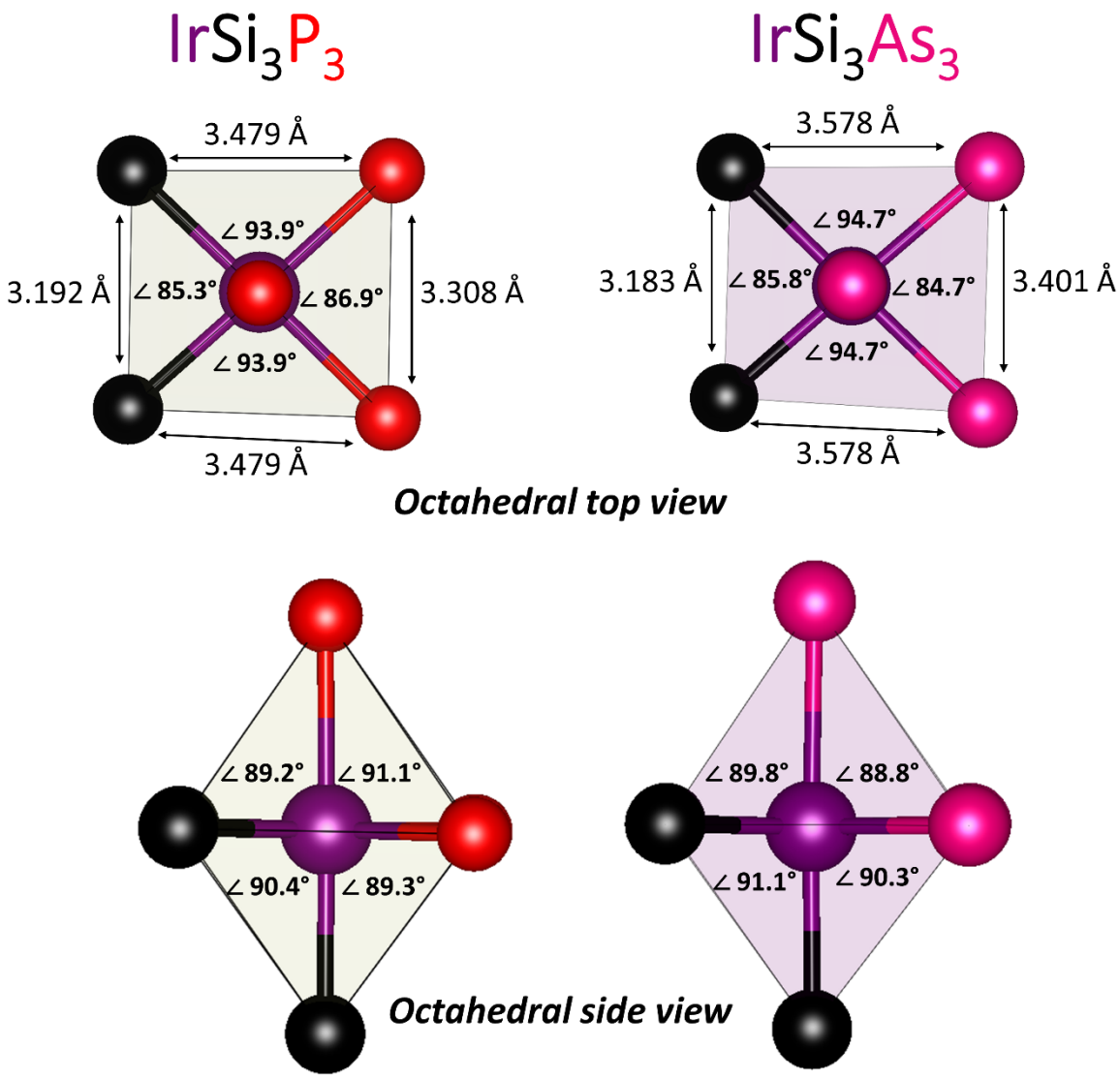
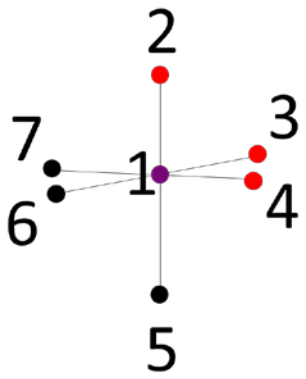
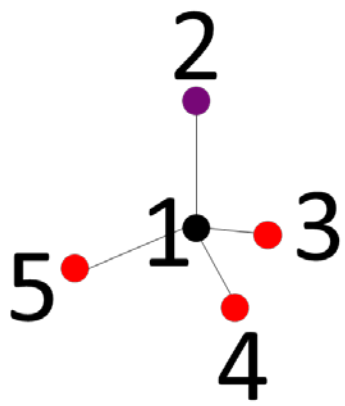


Figure S1. Distortions of octahedral building blocks compared for $IrSi_3P_3$ and $IrSi_3As_3$.

Table S1. Octahedral and tetrahedral angles and deviations from ideal geometry for $IrSi_3P_3$ and $IrSi_3As_3$ structures.

Angle Labels using legend below 	**Angle (°)**	**|Deviation from 90° |**
∠ 2-1-3 = ∠ 2-1-4:		
$IrSi_3P_3$	91.06(6)	1.06
$IrSi_3As_3$	88.8162(3)	1.18
∠ 2-1-6 = ∠ 2-1-7		
$IrSi_3P_3$	89.26(5)	0.74
$IrSi_3As_3$	89.7975(2)	0.20
∠ 5-1-3 = ∠ 5-1-4		
$IrSi_3P_3$	89.28(6)	0.72
$IrSi_3As_3$	90.2890(3)	0.29
∠ 5-1-6 = ∠ 5-1-7		
$IrSi_3P_3$	90.40(7)	0.40
$IrSi_3As_3$	91.0895(3)	1.09
∠ 3-1-4		
$IrSi_3P_3$	86.92(7)	3.08
$IrSi_3As_3$	84.720(1)	5.28
∠ 4-1-6 = ∠ 7-1-3		
$IrSi_3P_3$	93.90(5)	3.90
$IrSi_3As_3$	94.713(1)	4.71
∠ 6-1-7		
$IrSi_3P_3$	85.28(7)	4.72
$IrSi_3As_3$	85.820(1)	4.18
	Sum of deviation of all angles	
	$IrSi_3P_3$	**21.44**
	$IrSi_3As_3$	**24.40**

Angle Labels using legend below 	Angle (°) Si1 centered | Si2 centered	|Deviation from 109.5° |
∠ 2-1-3		
$IrSi_3P_3$	116.47(7) | 110.34(9)	6.97 | 0.84
$IrSi_3As_3$	117.8097(7) | 111.8321(4)	8.31 | 2.33
∠ 2-1-4:		
$IrSi_3P_3$	109.64(7) | 128.12(1)	0.14 | 18.62
$IrSi_3As_3$	111.0572(8) | 129.5553(9)	1.56 | 20.06
∠ 2-1-5:		
$IrSi_3P_3$	129.43(8) | 110.34(9)	19.93 | 0.84
$IrSi_3As_3$	130.1373(7) | 111.8321(4)	20.65 | 2.33
∠ 3-1-4		
$IrSi_3P_3$	105.67(8) | 94.42(1)	3.83 | 15.08
$IrSi_3As_3$	103.1513(5) | 93.0229(9)	6.35 | 16.48
∠ 4-1-5		
$IrSi_3P_3$	96.92(9) | 94.42(1)	12.58 | 15.08
$IrSi_3As_3$	95.4400(7) | 93.0229(9)	14.06 | 16.48
∠ 5-1-3		
$IrSi_3P_3$	95.24(6) | 118.90(1)	14.26 | 9.40
$IrSi_3As_3$	94.535(1) | 116.1872(7)	14.97 | 6.69
	Sum of deviation of all angles	
	$IrSi_3P_3$	**117.57**
	$IrSi_3As_3$	**130.27**

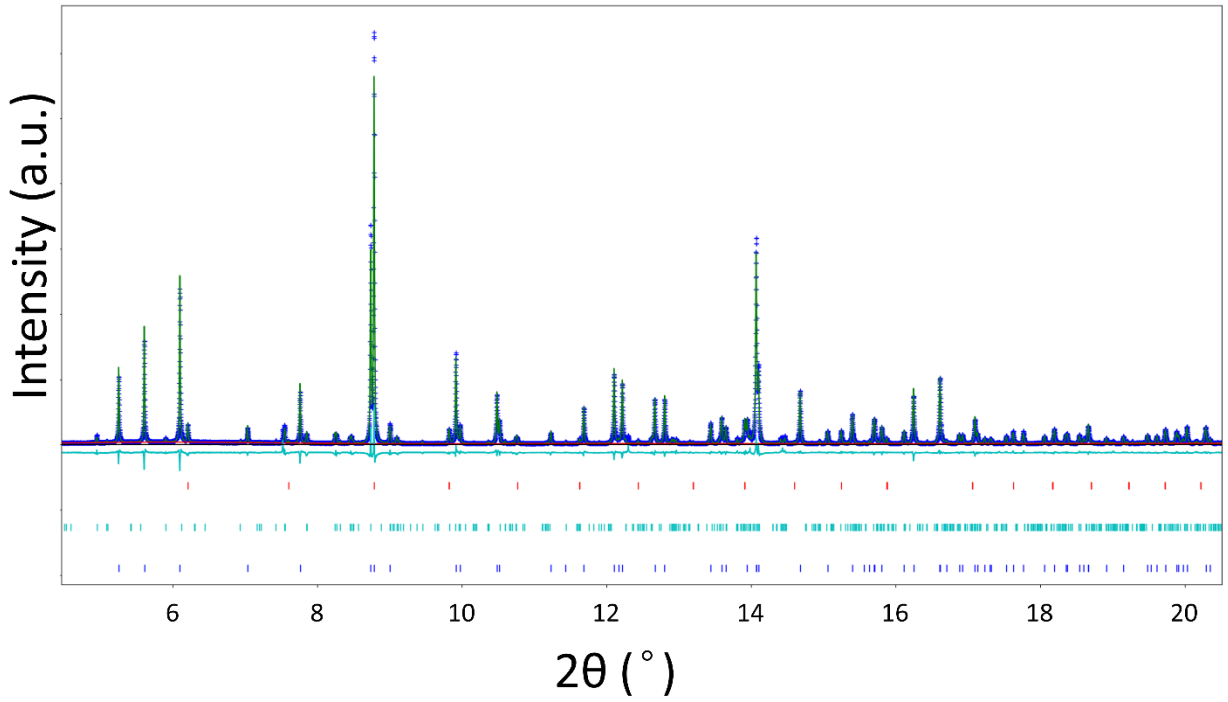


Figure S2. Rietveld refinement with main phase $IrSi_3As_3$ (phase fraction (p.f.) of 0.96; shown as dark blue tick marks), and admixture phases $IrAs_3$ (p.f. 0.02; red tick marks) and Ir_3Si_5 (p.f. 0.02; light blue tick marks) included. Difference curve shown in light blue, χ^2 = 2.0, *wR* = 10.7%.

Table S2. Atomic positions of Rietveld refined $IrSi_3As_3$ structural model with values in parentheses from the original single crystal X-ray diffraction structural solution.

Atom label	Wyckoff label	*x/a*	*y/b*	*z/c*	U_{iso}
Ir	2*a*	0.18447 (0.18633)	0	0.21214 (0.21623)	0.0004 (0.0013)
Si1	4*b*	0.61772 (0.61840)	0.28712 (0.28840)	0.88150 (0.88420)	0.0025 (0.0022)
Si2	2*a*	0.57094 (0.57240)	0	0.34589 (0.35160)	0.0025 (0.0036)
As1	2*a*	0.75670 (0.75910)	0	0.07415 (0.07930)	0.0011 (0.0003)
As2	4*b*	0.74726 (0.74850)	0.27258 (0.27197)	0.56891 (0.57180)	0.0011 (0.0008)

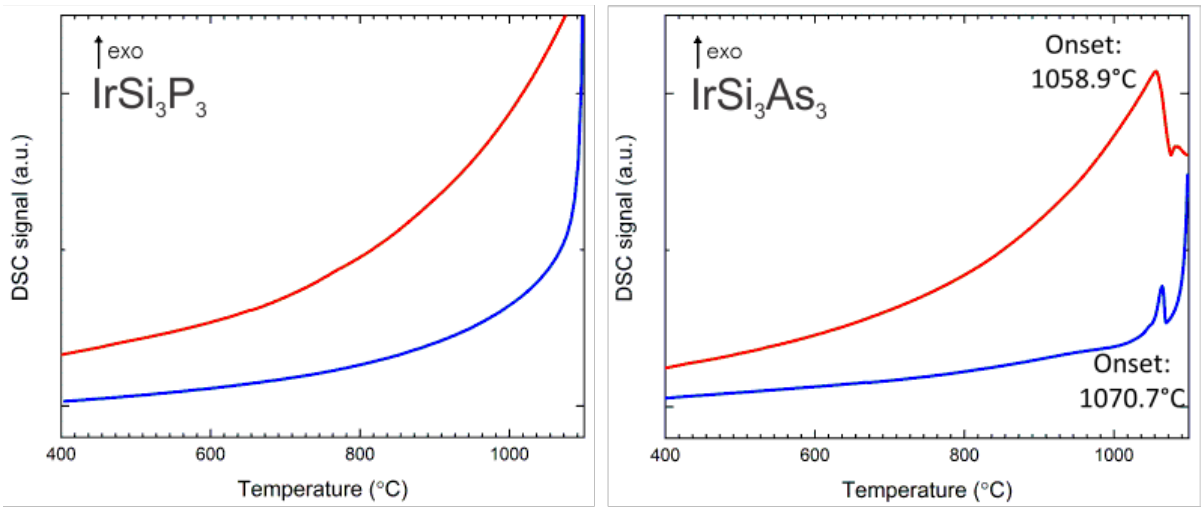


Figure S3. Differential scanning calorimetry plots of $IrSi_3P_3$ and $IrSi_3As_3$ with decomposition temperature onsets at 1059°C (upon heating – red curve) and 1071°C (upon cooling – blue curve).

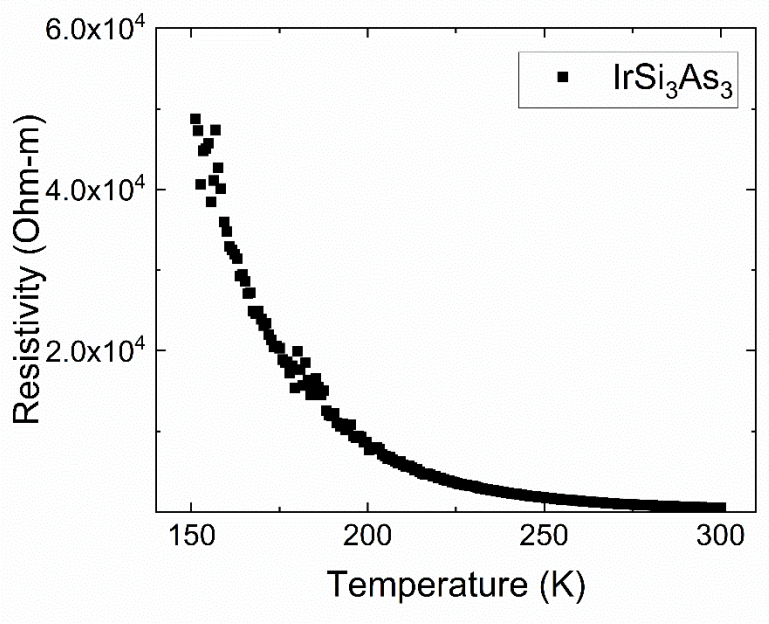


Figure S4. Electrical resistivity of sintered pellet of $IrSi_3As_3$ (150-300 K).