

Supporting Information

Polarization-resolved and helicity-resolved Raman spectra of monolayer XP_3 (X=Ge and In)

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Structural details for monolayer XP_3 (X=Ge, In)

Here is the POSCAR information for optimized GeP_3 monolayer.

----- POSCAR file -----

GeP3_mono

6.94096652600000

0.5002831386450043 -0.8665158144574282 0.0000000000000000

0.5002831386450043 0.8665158144574282 0.0000000000000000

0.0000000000000000 0.0000000000000000 4.3172736957890425

Ge P

2 6

Direct

0.3333333429999996 0.6666666870000029 0.9763144396388121

0.6666666269999979 0.3333333129999971 0.0036056361528162

0.8532868839873115 0.1467130860127082 0.9857248616266605

0.1670175341884261 0.8329825258115646 0.0257215953347332
0.8532869439873093 0.7065737679745625 0.9857248616266605
0.1670175041884307 0.3340350383768426 0.0257215953347332
0.2934262020254332 0.1467131160127036 0.9857248616266605
0.6659649316231511 0.8329824658115668 0.0257215953347332

Here is the POSCAR information for optimized InP₃ monolayer.

----- POSCAR file -----

InP3 mono

1.00114000000000
7.5571854270935299 -0.0000000000006986 0.0000000000000001
-3.7785927135980968 6.5447145610357662 0.0000000000000000
0.0000000000000002 -0.0000000000000005 29.9658389436042896

P In

6 2

Direct

0.5243906679417273 0.4756093320095371 0.0893529420716831
0.5243906679798479 0.0487813359189241 0.0893529420716831
0.9512186641145569 0.4756093320095371 0.0893529420716831
0.8089426455597709 0.1910573545092793 0.1303137263949821
0.8089426455216503 0.6178853505998924 0.1303137263949821
0.3821146791895980 0.1910573545092793 0.1303137263949821
0.0000000000000000 0.0000000000000000 0.1089165405801138
0.3333333432638810 0.6666666865188162 0.1107501278865515

Table SI Raman tensor elements a, b, c and d for monolayer GeP₃.

Mode	A ₁ (88)		A ₁ (93)		A ₁ (314)		A ₁ (327)		A ₁ (448)	
Laser(nm)	633	532	633	532	633	532	633	532	633	532
a(arb. units)	32e ^{-i×25°}	80 e ^{i×56°}	35 e ^{-i×71°}	58 e ^{i×82°}	103 e ^{-i×68°}	43 e ^{-i×64°}	101 e ^{-i×15°}	57 e ^{i×85°}	197 e ^{-i×16°}	190 e ^{-i×10°}
b(arb. units)	13 e ^{-i×2°}	12 e ^{-i×0°}	21 e ^{i×4°}	16 e ^{i×8°}	36 e ^{-i×2°}	26 e ^{i×2°}	27 e ^{-i×7°}	25 e ^{i×7°}	92 e ^{-i×4°}	78 e ^{i×4°}
Mode	E(83)		E(97)		E(127)		E(268)		E(329)	
Laser(nm)	633	532	633	532	633	532	633	532	633	532
c(arb. units)	17 e ^{-i×69°}	11 e ^{i×68°}	24 e ^{i×40°}	17 e ^{i×34°}	36 e ^{-i×53°}	11 e ^{i×38°}	30 e ^{-i×30°}	7 e ^{-i×45°}	35 e ^{i×13°}	41 e ^{i×67°}
d(arb. units)	4 e ^{i×52°}	3 e ^{-i×75°}	4 e ^{-i×56°}	6 e ^{i×12°}	2 e ^{i×23°}	6 e ^{-i×50°}	24 e ^{i×7°}	38 e ^{i×36°}	3 e ^{-i×60°}	6 e ^{-i×60°}
Mode	E(412)		E(435)							
Laser(nm)	633	532	633	532						
c(arb. units)	33 e ^{i×3°}	50 e ^{i×82°}	53 e ^{i×35°}	74 e ^{i×66°}						
d(arb. units)	20 e ^{i×5°}	29 e ^{i×6°}	22 e ^{i×30°}	13 e ^{i×32°}						

Table SII Raman tensor elements e, f, g and h for monolayer InP₃.

Mode	A _{1g} (64)		A _{1g} (281)		A _{1g} (452)					
Laser(nm)	633	532	633	532	633	532				
e(arb. units)	18 e ^{-i×82°}	35 e ^{-i×133°}	171 e ^{i×150°}	186 e ^{i×155°}	119 e ^{-i×26°}	119 e ^{i×3°}				
f(arb. units)	7 e ^{i×178°}	4 e ^{-i×63°}	2 e ^{i×63°}	11 e ^{-i×25°}	13 e ^{-i×8°}	25 e ^{-i×16°}				
Mode	E _g (68)		E _g (78)		E _g (275)		E _g (413)			
Laser(nm)	633	532	633	532	633	532	633	532	633	532
g(arb. units)	24 e ^{-i×15°}	16 e ^{-i×4°}	56 e ^{-i×44°}	41 e ^{-i×6°}	76 e ^{-i×25°}	63 e ^{i×11°}	92 e ^{-i×160°}	73 e ^{-i×105°}		
h(arb. units)	1 e ^{-i×112°}	2 e ^{-i×51°}	1 e ^{i×57°}	2 e ^{i×65°}	9 e ^{-i×120°}	11 e ^{-i×78°}	16 e ^{i×11°}	21 e ^{i×20°}		

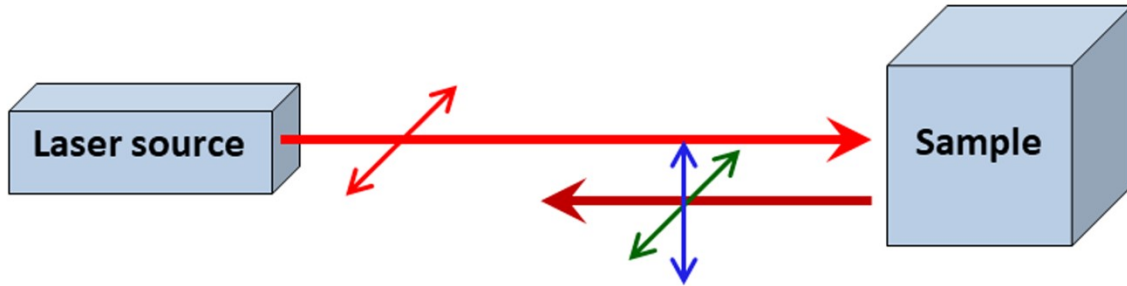


Figure S1 Schematic of the optical setup for polarization-resolved Raman spectroscopy. The polarization angle of the incident laser is changed by rotating the sample. The detection polarization of scattered light is kept parallel or perpendicular to the incident polarization. The optical setup for helicity-resolved Raman scattering measurements is similar but the samples are excited with circularly polarized light. The helicity of incident light is fixed and the helicity of Raman scattered light is selected to be the same or opposite to the incident light.

Table SIII Helicity-resolved Raman intensities of Raman active modes for monolayer GeP₃ when the wave vector k of incident light is along the y- and the z-axis.

k	Config	Laser (nm)	mode											
			E(83)	E(97)	E(127)	E(268)	E(329)	E(412)	E(435)	A ₁ (88)	A ₁ (93)	A ₁ (314)	A ₁ (327)	A ₁ (448)
y-axis	I ^{co} (arb. units)	633	0.73	1.02	1.38	0.22	0.20	0.12	0.28	4.34	3.99	2.74	2.79	7.85
		532	0.59	1.00	0.24	0.02	0.56	0.55	1.11	33.01	16.20	1.25	1.51	13.39
	I ^{cross} (arb. units)	633	0.91	1.15	1.40	0.79	0.21	0.29	0.47	0.91	2.54	1.61	0.93	1.13
		532	0.73	1.46	0.55	2.88	0.62	1.28	1.23	23.53	12.09	0.59	1.12	2.56
z-axis	I ^{co} (arb. units)	633	0.00	0.00	0.00	0.00	0.00	0.00	0.00	8.94	9.59	7.76	6.92	14.78
		532	0.00	0.00	0.00	0.00	0.00	0.00	0.00	110.50	52.39	2.72	4.41	27.32
	I ^{cross} (arb. units)	633	5.83	8.18	11.00	1.77	1.64	0.98	2.27	0.00	0.00	0.00	0.00	0.00
		532	4.68	7.98	1.89	0.19	4.51	4.41	8.84	0.00	0.00	0.00	0.00	0.00

Table SIV Helicity-resolved Raman intensities of Raman active modes for monolayer InP₃ when the wave vector k of incident light is along the y- and the z-axis.

k	Config	Laser(nm)	mode						
			E _g (68)	E _g (78)	E _g (275)	E _g (413)	A _{1g} (64)	A _{1g} (281)	A _{1g} (452)
y-axis	I ^{co} (arb. units)	633	0.21	0.86	0.14	0.09	0.13	0.66	0.16
		532	0.18	0.92	0.19	0.12	1.09	1.39	0.38
	I ^{cross} (arb. units)	633	0.21	0.86	0.14	0.10	0.17	0.66	0.11
		532	0.20	0.93	0.21	0.16	0.93	1.76	0.17
z-axis	I ^{co} (arb. units)	633	0.00	0.00	0.00	0.00	0.53	2.65	0.53
		532	0.00	0.00	0.00	0.00	3.99	6.26	1.06
	I ^{cross} (arb. units)	633	1.66	6.85	1.10	0.74	0.00	0.00	0.00
		532	1.47	7.33	1.50	0.94	0.00	0.00	0.00