Electronic Supplementary Material (ESI) for Nanoscale.

Supplemental Material

One-dimensional metal thiophosphate nanowires by cluster assembling

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Table S1 Band gap (in the unit of eV) of 1D, 2D and bulk MPS_4 calculated by different methods, including the PBE functional, PBE+*U* method (*U* = 3.0 eV), and HSE06 hybrid functional. For ferromagnets, the band gap of spin-up and spin-down channels are given.

	1D		bulk	
	PBE	HSE06	PBE	HSE06
BPS_4	2.37	3.57	1.97	3.21
$AlPS_4$	2.93	4.14	2.39	3.55
$GaPS_4$	2.78	4.01	2.02	3.17
$InPS_4$	2.53	3.73	1.99	3.12

	1D		2D	
	PBE	PBE+U	PBE	PBE+U
VPS_4	0.77	0.60	0.01	0.03
$CrPS_4$	0.97	1.15	1.00/1.41	0.66/1.69
$CoPS_4$	0.76	0.81	0.87	0.71
$NiPS_4$	1.73/0.01	1.73/0.22	--	--
$MoPS_4$	--	--	--	--
$RuPS_4$	--	--	0.33	0.37
$PdPS_4$	0	0	--	--
$RePS_4$	0.02/metal	1.41/metal	--	--

1D	$ScPS_4$	$TiPS_4$	$MnPS_4$	$FePS_4$	YPS_4	$ZrPS_4$	$NbPS_4$	$RhPS_4$
PBE	2.07	--	1.25	0.66	2.32	--	0.91	0.64
PBE+*U*	2.28	--	1.56	1.65	0.37	--	1.01	0.83
	$HfPS_4$	$TaPS_4$	WPS_4	$OsPS_4$	$IrPS_4$	$PtPS_4$		
PBE	--	1.77	--	--	0.72	0		
PBE+*U*	--	1.73	--	--	0.94	0		

Table S2 Formation energies (E_{form}) of 1D MPS_4 (M = transitional metals) nanowires.

3*d*	E_{form} (eV)	4*d*	E_{form} (eV)	5*d*	E_{form} (eV)
$ScPS_4$	-3.77	YPS_4	-3.89	--	--
$TiPS_4$	-2.19	$ZrPS_4$	-2.43	$HfPS_4$	-2.45
VPS_4	-1.21	$NbPS_4$	-1.59	$TaPS_4$	-1.46
$CrPS_4$	-1.44	$MoPS_4$	0.13	WPS_4	0.63
$MnPS_4$	-1.15	--	--	$RePS_4$	1.06
$FePS_4$	-3.04	$RuPS_4$	0.69	$OsPS_4$	1.28
$CoPS_4$	-0.16	$RhPS_4$	0.25	$IrPS_4$	0.69
$NiPS_4$	-0.08	$PdPS_4$	0.06	$PtPS_4$	-0.10

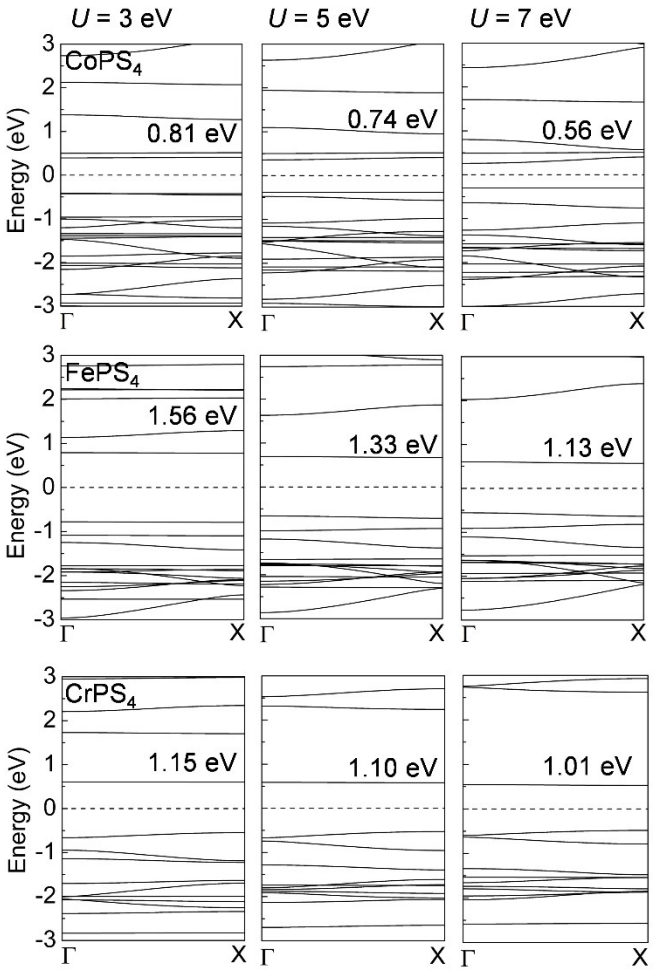


Fig. S1 Band structures of MPS_4 (M = Co, Fe and Cr) nanowires by PBE+U with U = 3.0 eV, U = 5.0 eV and U = 7.0 eV.

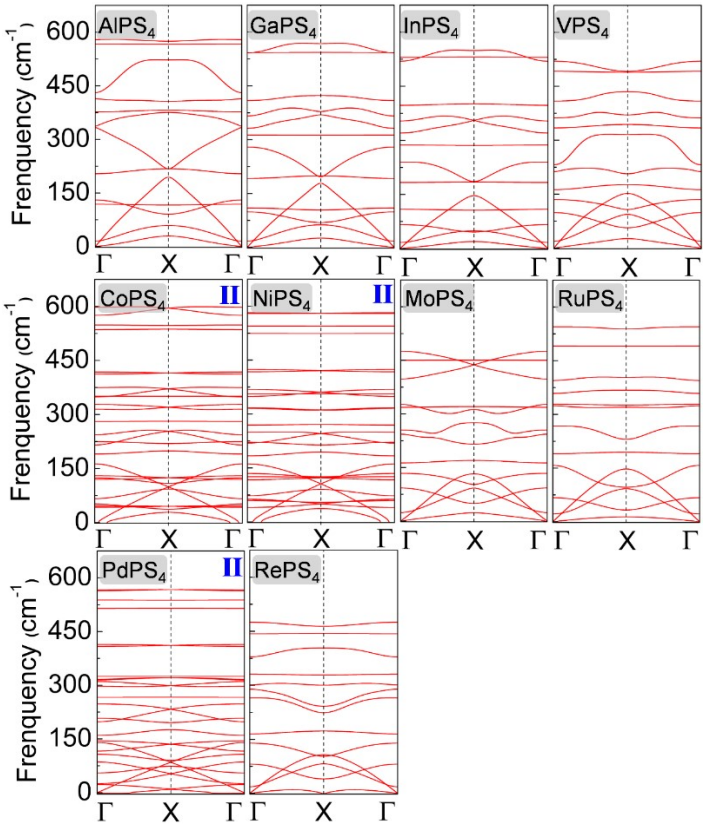


Fig. S2 Band structures of MPS_4 (M = Al, Ga, In, V, Co, Ni, Mo, Ru, Pd, and Re) nanowires.

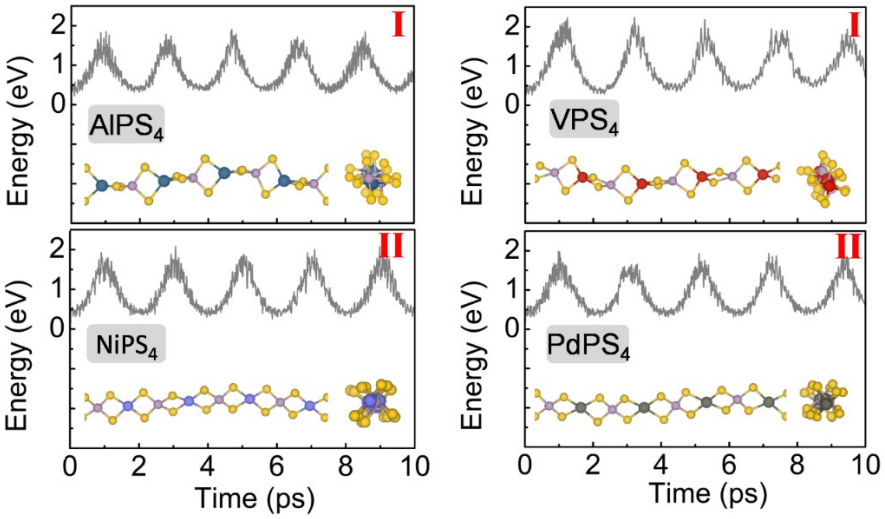


Fig. S3 Energy profile during AIMD simulation for MPS_4 (M = Al, V, Ni and Pd) nanowires at 300 K. The energy is referred to that of the equilibrium structure of MPS_4 nanowire at 0 K.

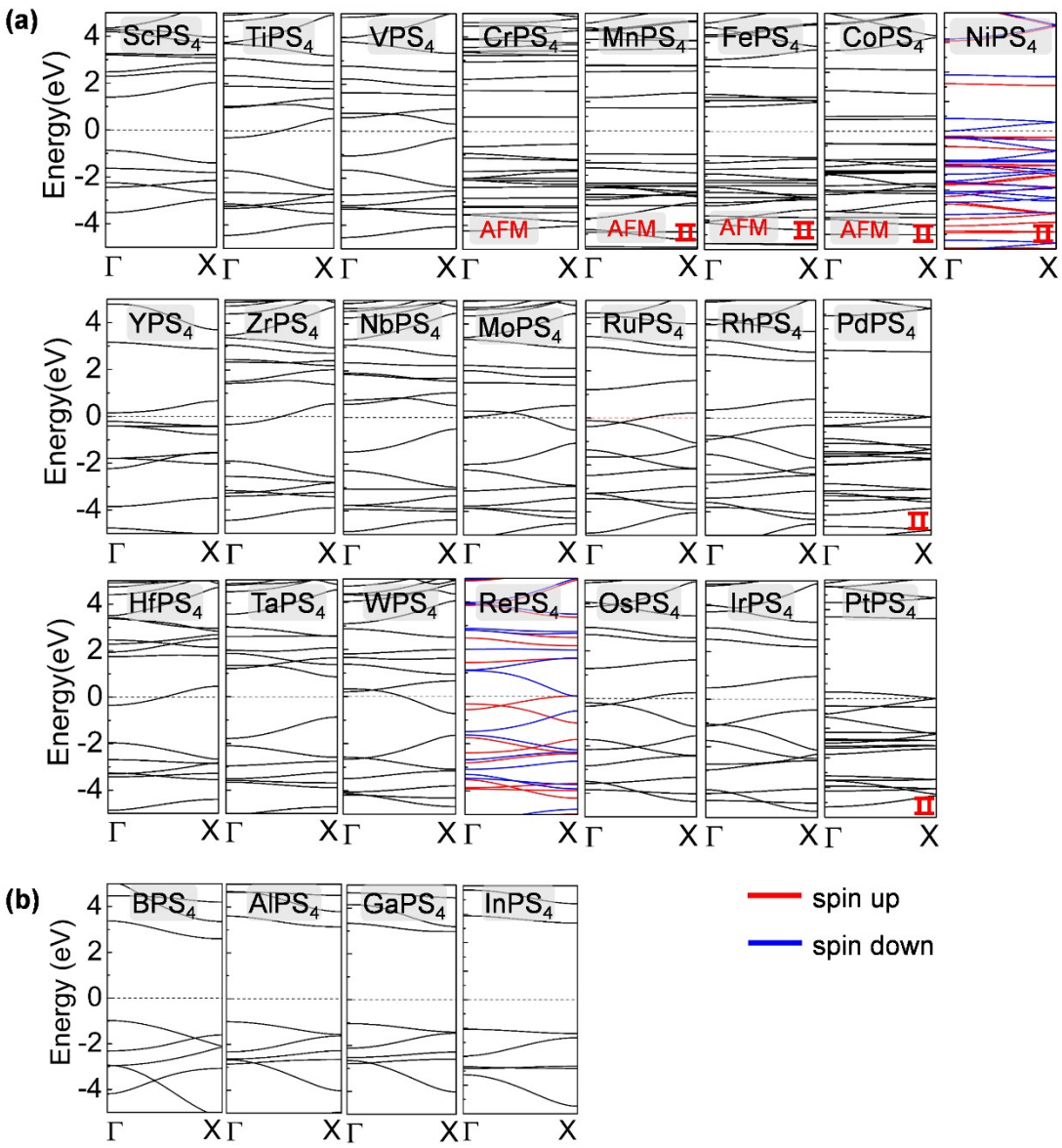


Fig. S4 Band structures of MPS_4 nanowires for (a) M = transition metal elements, calculated by the PBE+*U* (*U* = 3.0 eV) method, and (b) M = group-IIIA elements (B, Al, Ga, In), calculated by the HSE06 functional. The symbol "II" indicate the type-II structure for the nanowire.

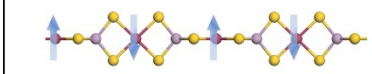
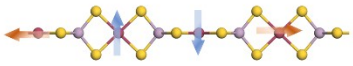
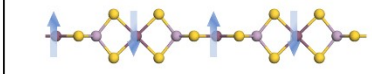
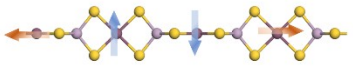
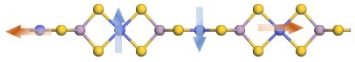
	collinear	non-collinear	*E* (meV)
$MnPS_4$			4.8
$FePS_4$			2.0
$CoPS_4$			1.6

Fig. S5 The lowest-energy collinear magnetic order and non-collinear magnetic configurations of selected MPS_4 nanowires. MPS_4 (M = Mn, Fe, and Co) nanowires are found to have non-collinear magnetic order, with higher energy by 1.6 ~ 4.8 meV per magnetic atom than those of the collinear ones.

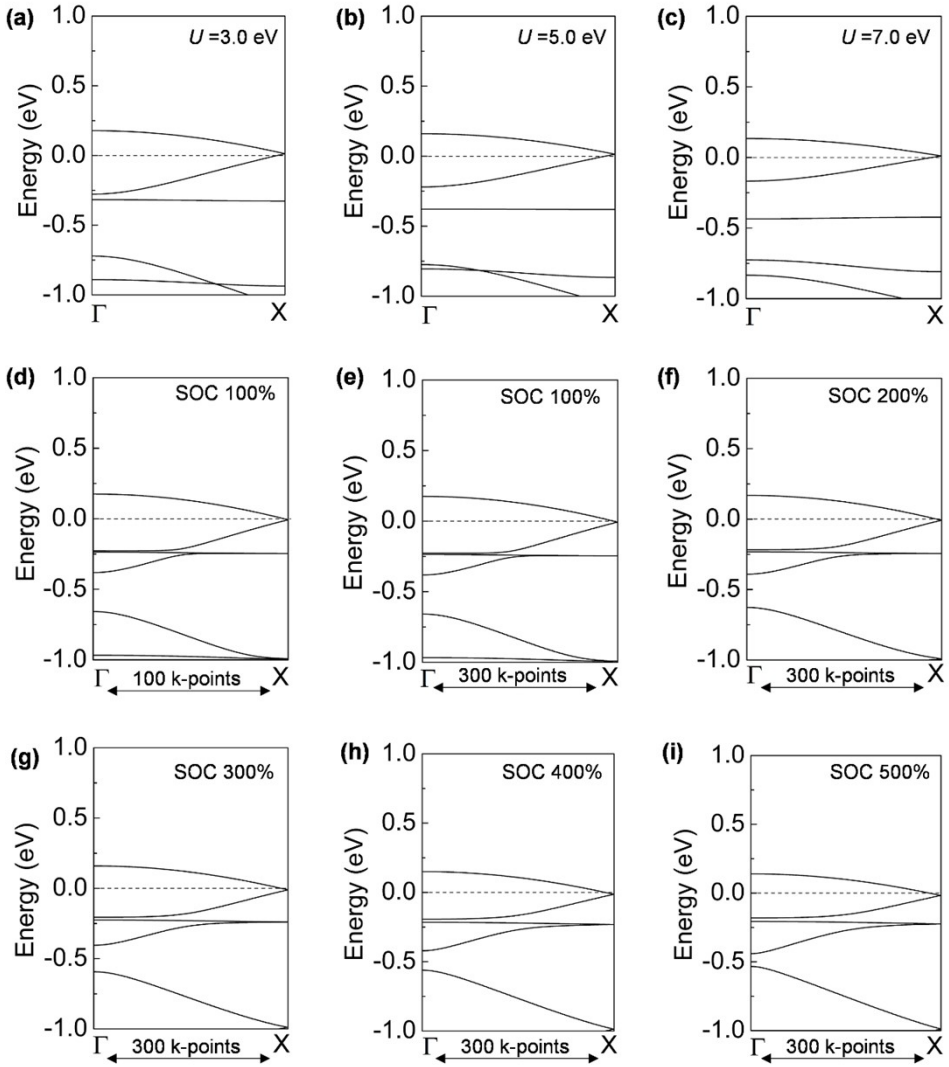


Fig. S6 Band structures of $PdPS_4$ nanowire calculated by different methods, (a) PBE+U with U = 3.0 eV, (b) U = 5.0 eV, (c) U = 7.0 eV, (d) by considering SOC with different densities of **k**-points with 0.0002 $Å^{-1}$, and (f) ~ (i) by considering different SOC strength (200% ~ 500%) and denser **k**-points with 0.0001 $Å^{-1}$. The semimetal behavior is robust under different computational methods.

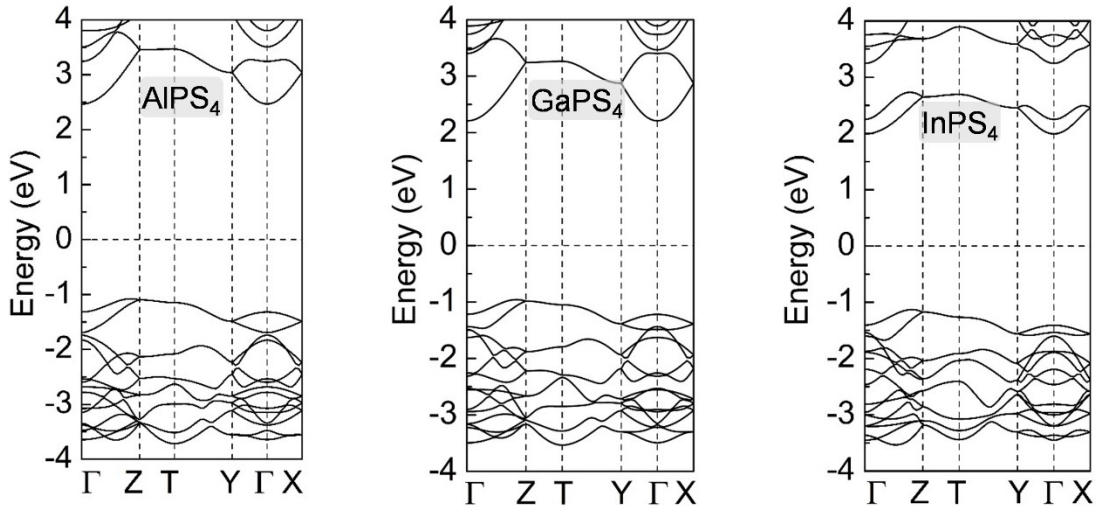


Fig. S7 Band structures of bulk MPS_4 (M = Al, Ga, In) calculated by the HSE06 functional.

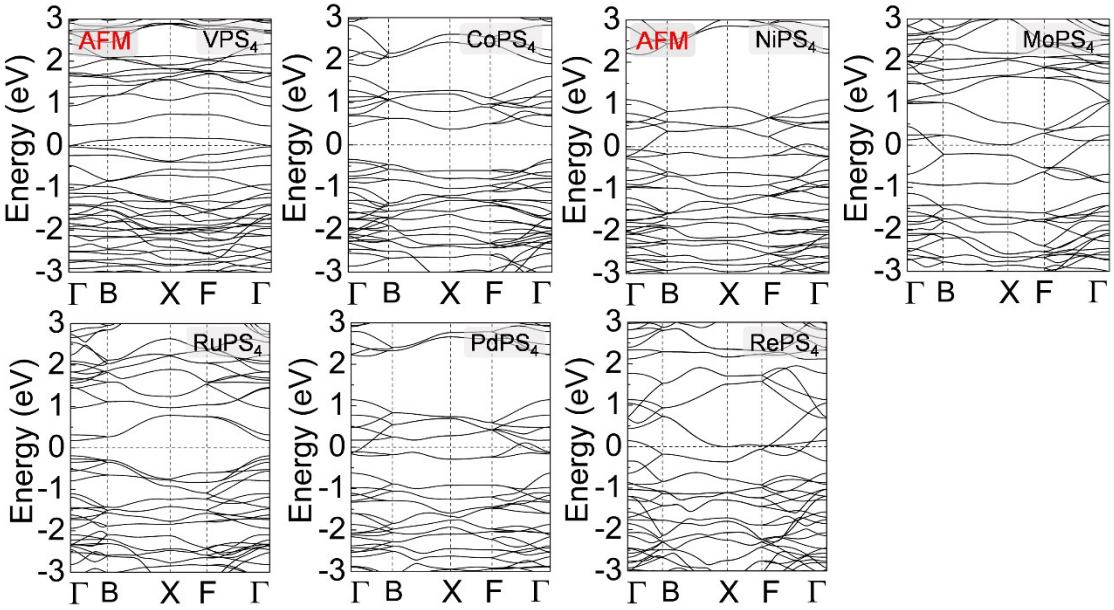


Fig. S8 Band structures of 2D MPS_4 (M = V, Co, Ni, Mo, Ru, Pd, Re) monolayer calculated by the PBE+ U (U = 3.0 eV) method.