AMP^2: a fully automated program for ab initio calculations of crystalline materials

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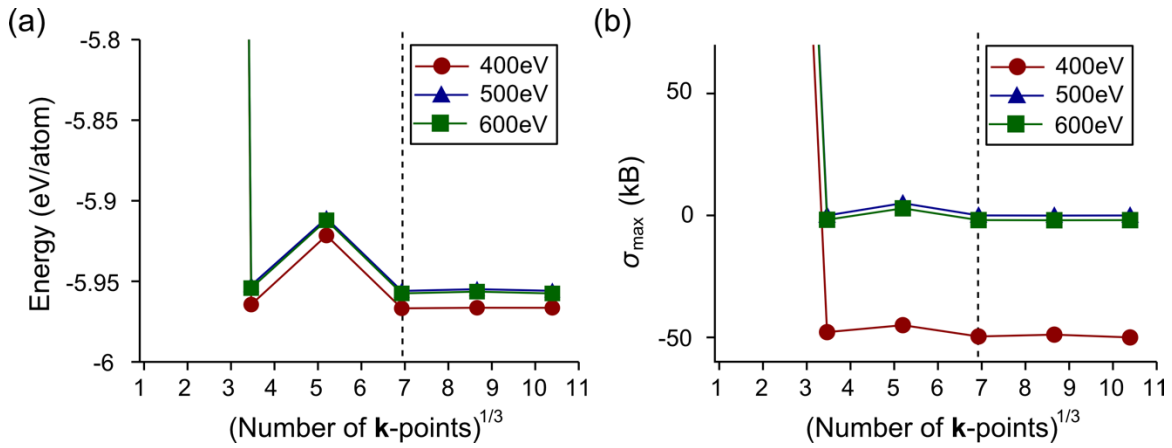

Figure S1 (a) Energy and (b) σ_{max} convergence test of **k**-points for MgO. The vertical dashed lines indicate the converged **k**-points. The convergence of the energy (a) and pressure (b) for MgO with respect to the k-point grid, evaluated at the cutoff energy of 400, 500, and 600 eV. (400 eV is the default value of VASP while AMP^2 gives 500 eV for the converged E_{cut}.) It is seen that the convergence behavior of k-point is similar among different cutoff energies except for rigid shifts, meaning that the converged k-point grid can be safely determined at the default cutoff energy (as is currently done in AMP^2).

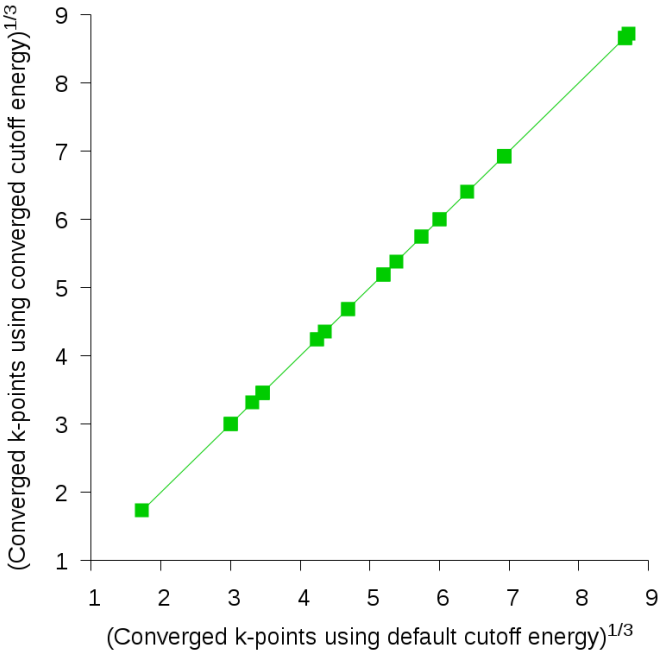

Figure S2 Converged k-point grids for default and converged cutoff energies for 30 randomly chosen materials. (BaSn, BrCs, CuCl, CaH_2, $CaNi_3$, H_2Sr, La_3Sn_7, $Li_{13}Sn_5$, Mn_3Sn, P_3N_5, $NaTaO_3$, SiO_2, ZnS, Tl_2S_3, $Ga_{10}La_2Ni$, GeLaRu, GePtSe, GeRhSc, IrI_3Te_6, $InTaS_2$, In_2PbS_4, $Ir_2Y_2O_7$, KLiSe, $KNbO_3$, K_2SnTe_3, $NaNbN_2$, $Na_6Sn_2Se_7$, Nb_4SiTe_4, $NiPSe_3$, and $PdSc_6Te_2$) They show perfect agreements. Therefore, we conclude that the converged k-point grid is insensitive to the cutoff energy and the convergence test can be carried out independently with respect to the k point and cutoff energy.

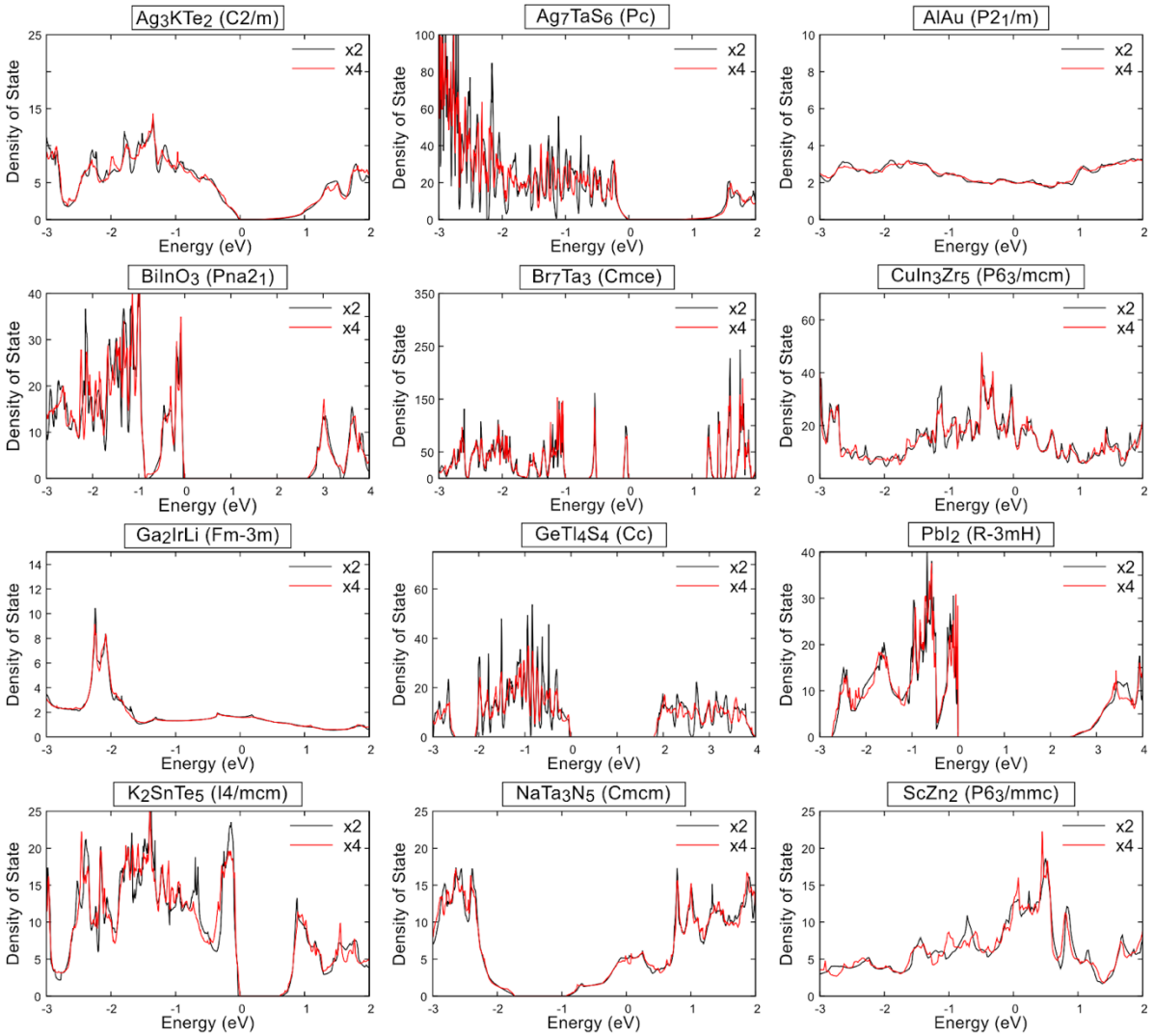

Figure S3 Density of states of Ag_3KTe_2, Ag_7TaS_6, AlAu, $BiInO_3$, Br_7Ta_3, $CuIn_3Zr_5$, Ga_2IrLi, $GeTl_4S_4$, PbI_2, K_2SnTe_5, $NaTa_3N_5$, and $ScZn_2$ with the different **k**-point grids. As shown in the figure, k-point doubling is sufficient for DOS figures.

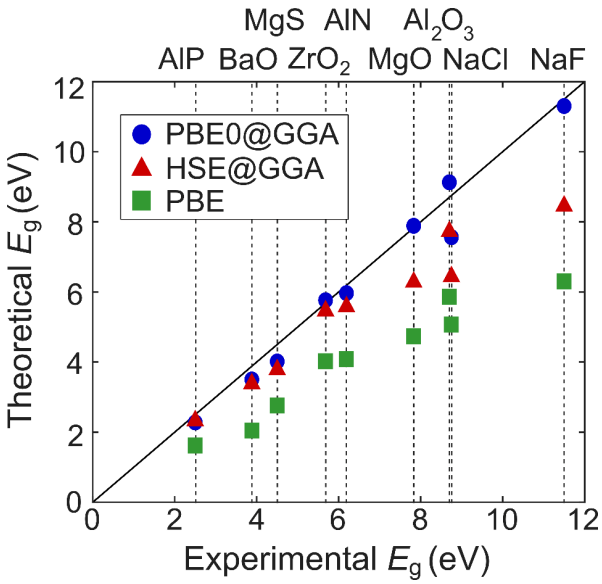

Figure S4 Comparison of theoretical and experimental band gap.[1–6] The solid line indicates perfect agreement with experiment.

Tests for gap-corrected band structure

For materials whose conduction band and valence bands are separated or touch each other (ex. Ag_2O, InAs, Ge, and InSb) within GGA, AMP^2 consistently produced get-corrected band structures as guaranteed by the proposed algorithm. When conduction and valence bands overlap each other, AMP^2 worked well for PdO, PtO, Pb_2PtO_4, and Sn_3O_4 but failed to separate them in Cd_3As_2 and $CaPd_3O_4$. While it is difficult to specify and quantify a feature in band structures that splits success versus failure, we find that GGA and HSE band structures are significantly different in the two materials. For example, the figure below compares GGA and HSE band structures of $CaPd_3O_4$, respectively. Based on the HSE result, one can visually identify in the GGA band structure the conduction and valence bands that should move up and down upon gap correction, as indicated by red and blue lines, respectively. One can see that some parts of the lines are broken as indicated by the dashed lines, which is due to the orbital hybridization between conduction and valence bands. This is in contrast with Fig. 5 in the manuscript in which the hybridization is weak. Therefore, the proposed scheme would fail because orbital characters are highly mixed in the broken regions.

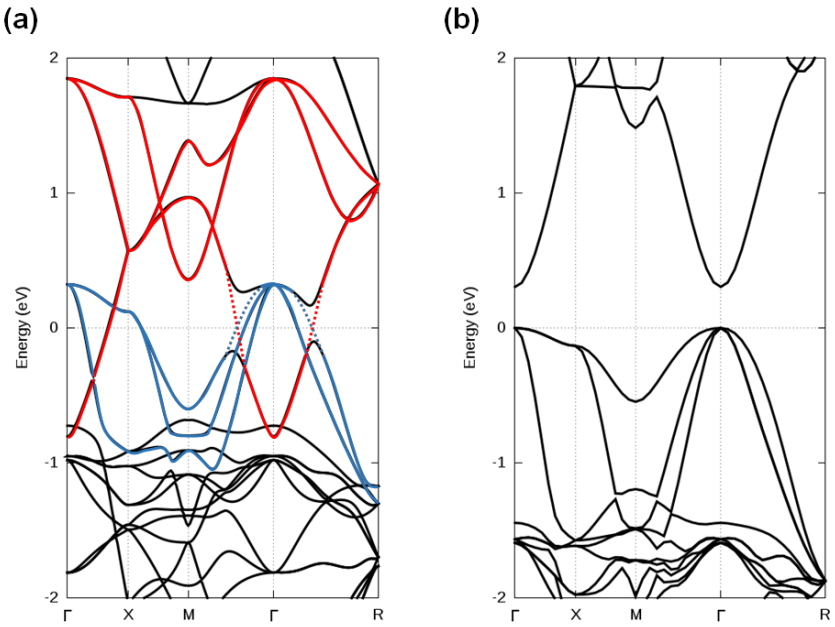

Figure S5 Band structure of $CaPd_3O_4$ using (a) GGA, (b) HSE functionals, respectively.

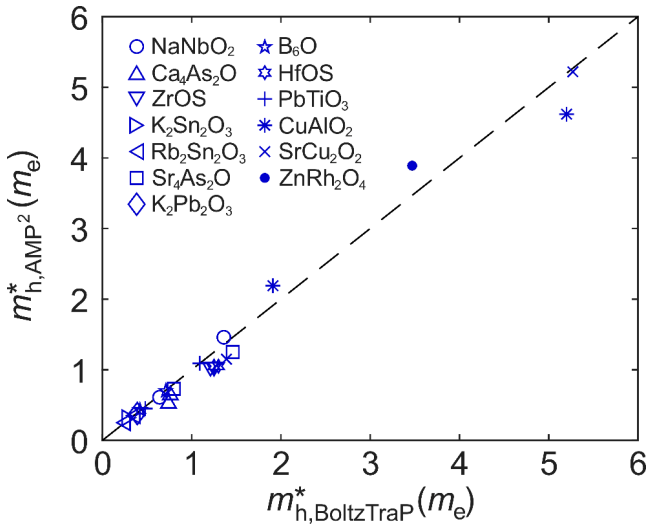

Figure S6 Comparison of principle components of hole effective mass tensor between AMP[2] and Boltztrap.[7] The solid line indicates perfect agreement.

Table S1. The list and success/failure of 100 materials.

Chemical formula (ICSD number)

Materials with successful termination (86)					
$AgAsSe_2$ (20087)	CoGeNb (623540)	K_2CuF_4 (15372)	LuN (236553)	$RbInTe_2$ (75346)	VPO_5 (108983)
AgBr (52246)	$Cr(Te_2Rh)_2$ (626579)	K_2SnBr_6 (35557)	$LuTlS_2$ (642581)	RbSb (280591)	YZn_3 (106227)
$AlCs_6Sb_3$ (300128)	CS_2 (15672)	KC_8 (70020)	Mg_3Au (58541)	$Sc(P_2Rh_3)_2$ (182778)	Y_4GaCo_4 (20953)
$AlFe_2Ni$ (57808)	Cs_2PtI_6 (201309)	KCd_4As_3 (262032)	MgB_2 (108064)	$ScInCu_2$ (103009)	Y_5S_7 (43620)
$AsPd_5$ (44042)	Cu_3Sb (44479)	$La(Ge_3Pt)_4$ (174552)	$MnPd_2$ (247774)	SiC (24630)	YIr_2 (104601)
B_2Mo_2Ir (23786)	$FeSi_2$ (24360)	$La(Sb_3Ru)_4$ (641783)	Na_2BiAu (261790)	SrF_2 (262349)	$YSnPd_2$ (105699)
$BaNiO_2$ (15760)	Ga_2Se_3 (35028)	LaCdPd (656599)	Na_2HgPb (261791)	$SrIr_2$ (104564)	ZnO (190802)
$BeBr_2$ (92584)	Ga_3Fe (631756)	$LaGa_2$ (103766)	$NaMoF_6$ (27484)	$SrMnBi_2$ (100025)	Zr_5Sb_3Ru (650598)
BiI (1559)	Ge_2Mo (16822)	Li_2Si (24146)	NaN_3 (644523)	Ta_2N (185289)	ZrAg (58390)
$Ca(InP)_2$ (260562)	Hf_2SN_2 (250915)	Li_6Br_3N (16798)	$Nb_7(BC)_4$ (411624)	Ta_5Ge_3 (195542)	$ZrNi_3$ (105482)
$Ca(MnAs)_2$ (41792)	$Hf_3B_2Ir_5$ (44316)	$LiAg_2Sn$ (151446)	PbI_2 (108906)	TaRu (650690)	ZrSiRu (16306)
Ca_2Ag_7 (55510)	$HfFeCl_6$ (39817)	$LiGa_2Pt$ (106717)	$PdCl_2$ (421220)	TaSiRh (90435)	
$Ca_3Al_7Ag_2$ (104173)	Hg_2Rh (106786)	Lu_2ReC_2 (618232)	PtO_2 (77654)	TeIr (44870)	
$CaSb_2$ (862)	HgCl (157979)	Lu_3Al_2 (57959)	Rb_2HgF_4 (72352)	Ti_4P_3 (648219)	
CdTe (246692)	ICI (23886)	$LuIn_3$ (51969)	Rb_2TiCl_6 (26689)	Tl_2ZnI_4 (37099)	

Material with any kinds of errors (14)			
Material	**Stage in error**	**Reason**	**Category**
$BaCoS_2$ (82639)	Magnetic ordering	Not converge	Termination
Cl_3NW (165376)	Effective mass	Memory	Crash
$Cr(Mo_3Se_4)_2$ (626324)	Kptest_w/o U	Not converge	Termination
Cr_2GaC (419116)	Kptest_w/o U	Not converge	Termination
$CsTiBr_3$ (400053)	Magnetic ordering	EDDDAV	Crash
Fe_4O_5 (185514)	Magnetic ordering	Not converge	Termination
Mn_2Au (58548)	Magnetic ordering	Not converge	Termination
Mn_3As (76409)	Magnetic ordering	Not converge	Termination
Ni_2PdSe_2 (419760)	Magnetic ordering	Not converge	Termination
NiF_3 (87943)	Effective mass	Memory	Crash
Sr_3GeO (413385)	HSE	Memory	Crash
V_5Ge_3 (44504)	Magnetic ordering	Not converge	Termination
VS_2 (68713)	Kptest	Not converge	Termination
$Y(Fe_2Si)_2$ (186048)	Magnetic ordering	Not converge	Termination

When we briefly checked for the magnetic ordering from Ising models, every computational result was reasonable except for $FeHfCl_6$ and K_2CuF_4 whose spin ordering are mistaken. $FeHfCl_6$ was reported as antiferromagnetic materials but it has ferromagnetic spin-ordering in AMP^2.[8] This is because the antiferromagnetic coupling between Fe ions exceeds the present cutoff of 5 Å. When we increased the cutoff distance, AMP^2 correctly predicts the antiferromagnetic spin ordering. However, energy difference is almost negligible (only 0.4 meV/Fe atom). On the other hand, K_2CuF_4 is a well-known two-dimensional ferromagnetic material but AMP^2 predicts an antiferromagnetic ordering.[9,10] It is known that the ferromagnetic order is more stable only if Jahn-Teller distortion is taken into account but the structure from ICSD is symmetric without Jahn-Teller distortions. When the symmetry is broken, we find that AMP^2 produces correct results.

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