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Supporting Information for

# Controlling the magnetic anisotropy of $Ru_mIr_n$ (m+n=3) clusters using MgO(001) substrate

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**Table S1.** The calculated MAE (in meV) of some systems versus $U_{eff}$ value (in eV).

| $U_{EFF}$ (RU) | $U_{EFF}$ (IR) | $Ru_2Ir$ | $Ir_3$ | $Ru_3$@MgO | $Ru_2Ir$@MgO | $RuIr_2$@MgO | $Ir_3$@MgO |
|---|---|---|---|---|---|---|---|
| **1.42** | 1.34 | 7.97 | -12.77 | -1.77 | -4.64 | -24.07 | 20.23 |
| **1.92** | 1.84 | 4.70 | -10.36 | -2.30 | 4.32 | 0.17 | 21.09 |
| **2.42** | 2.34 | 4.07 | -8.18 | -2.86 | 3.93 | 4.18 | 20.56 |
| **2.92** | 2.84 | 2.73 | -3.77 | -2.57 | -1.55 | 15.50 | 21.79 |
| **3.42** | 3.34 | 0.92 | 3.23 | -2.22 | -4.29 | 12.91 | 22.26 |

**Table S2.** The calculated $M_S$ (in $\mu_B$) of some systems versus $U_{eff}$ value (in eV).

| $U_{EFF}$ (RU) | $U_{EFF}$ (IR) | $Ru_2Ir$ | $Ir_3$ | $Ru_3$@MgO | $Ru_2Ir$@MgO | $RuIr_2$@MgO | $Ir_3$@MgO |
|---|---|---|---|---|---|---|---|
| **1.42** | 1.34 | 5.000 | 1.000 | 8.000 | 5.000 | 6.000 | 3.000 |
| **1.92** | 1.84 | 5.000 | 1.000 | 8.000 | 7.000 | 6.000 | 3.000 |
| **2.42** | 2.34 | 5.000 | 1.000 | 8.000 | 7.000 | 6.000 | 3.000 |
| **2.92** | 2.84 | 5.000 | 1.002 | 8.000 | 7.000 | 6.000 | 3.000 |
| **3.42** | 3.34 | 5.000 | 1.000 | 8.000 | 5.000 | 4.000 | 3.000 |

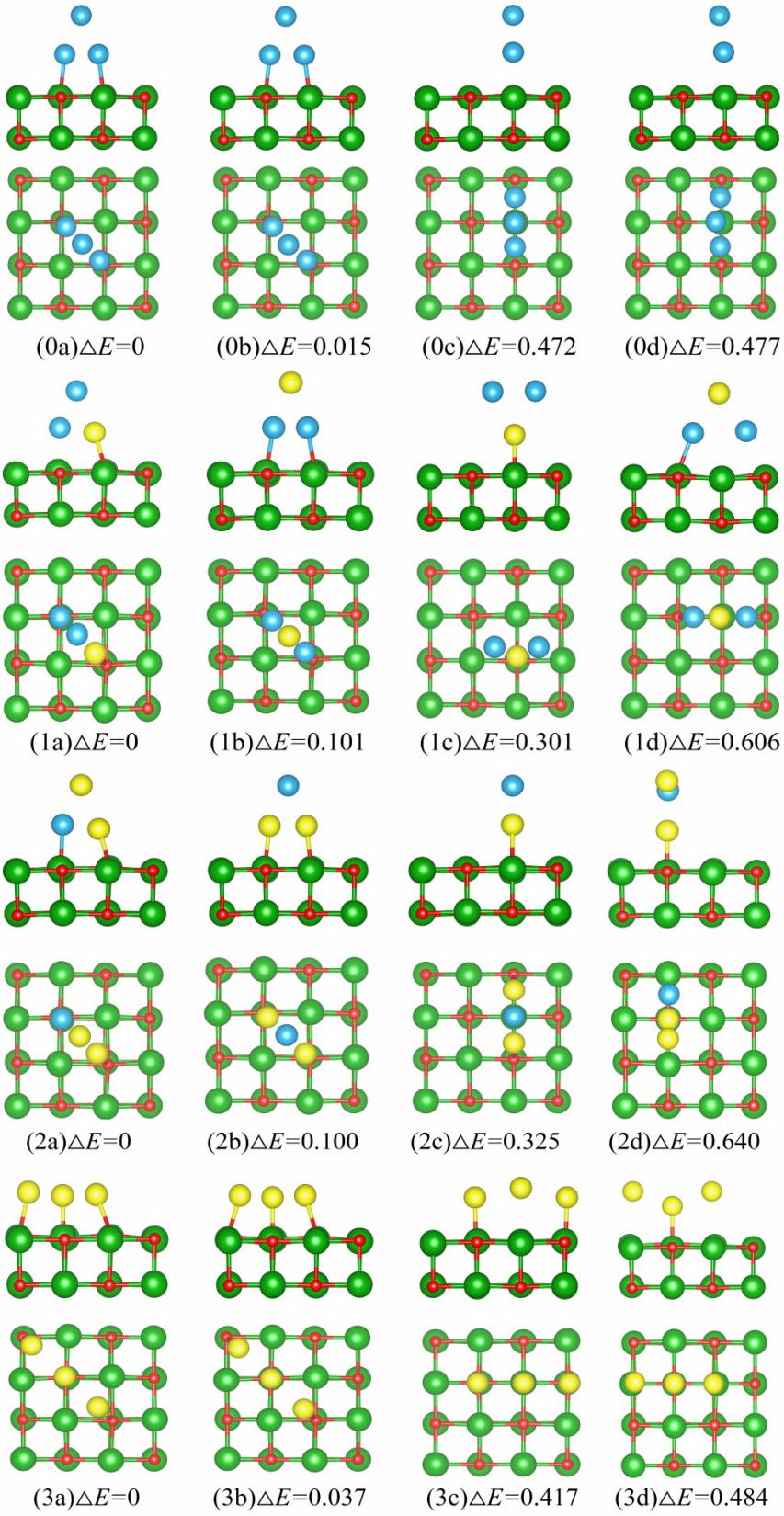


**Figure S1.** The energy differences between the sub-stable structure and the lowest energy structure of $Ru_mIr_n$@MgO. The numbers in brackets represent the number of Ir. Red, green yellow and blue balls represent Mg, O, Ir and Ru atoms, respectively.

**Table S3.** Coordinates of $Ru_mIr_n$ clusters and $Ru_mIr_n$@MgO (m+n=3) systems. All the lattice parameters of all periodic models are 8.4918 Å, 8.4918 Å and 22.1230 Å, respectively. The angles of α, β and γ is all 90°.

| STRUCTURE | COORDINATES |
|---|---|
| **$RU_3$** | Ru<br>3<br>Direct<br>0.1312046918880815 0.6129406256853983 0.3707849939842554<br>0.0328565744070551 0.7158917093293583 0.2858775583386560<br>0.2356687437048638 0.5128976929852423 0.2863674206770830 |
| **$RU_2IR$** | Ir Ru<br>1 2<br>Direct<br>0.4491740835586133 0.3049993311660446 0.2778575809786321<br>0.3491290684426694 0.4063521225895270 0.3711671152126086<br>0.2505268349987149 0.5033685052444281 0.2882453178087585 |
| **$RUIR_2$** | Ir Ru<br>2 1<br>Direct<br>0.4602058951413085 0.3138028744265862 0.2738622049359847<br>0.3659360534831258 0.4074868254772957 0.3708148570094618<br>0.2678680743755694 0.5057903010961137 0.2809729380545503 |

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|---|---|
| **$IR_3$** | Ir<br>3<br>Direct<br>0.4298753876607057 0.6817849697463595 0.2915016517828106<br>0.2482474474894239 0.5002665515639750 0.2920268973155439<br>0.0662571608498697 0.3191284706896841 0.2916714599016138 |

| STRUCTURE | COORDINATES |
|---|---|
| **$RU_3$@MGO** | O Mg Ru<br>16 16 3<br>Direct<br>0.0114966265655723 0.9883022642683736 0.0865538215648239<br>0.0125128848762091 0.4909985153269198 0.0889357530675204<br>0.2657534413221414 0.4849742310115945 0.1871295294881677<br>0.2619763118209845 0.9909339930374964 0.1841177994585077<br>0.2603819580000740 0.2407832531002037 0.0865925455649485<br>0.2594008721968509 0.7379333564321220 0.0889155164757287<br>0.5096070099865876 0.4900096888353394 0.0865750505384405<br>0.5103916436978145 0.9888458445775500 0.0852389472336582<br>0.5121079926939346 0.2384320955365311 0.1836680743698655<br>0.5123871508828435 0.7402995550711507 0.1841267336501753<br>0.7598784192798068 0.9905900054040043 0.1836393668092514<br>0.7616452903954581 0.2400917684702121 0.0852661520667926<br>0.7620837154524090 0.7388923067001034 0.0865867134946293<br>0.7595411146307635 0.4885304715950148 0.1841142009511783<br>0.0061473162173969 0.7443900415989518 0.1868108745582341<br>0.0102512275777265 0.2380223642523679 0.1841145326068835<br>0.0113442612779567 0.2398017034860452 0.0890417699131417<br>0.0111512295750015 0.7392805359585716 0.0896035311662178<br>0.2606483210531525 0.9891820798709416 0.0890608922249124<br>0.2688290909107531 0.7473963881207843 0.1858277762424736<br>0.2628370416514892 0.2336035548181321 0.1833490113215752<br>0.2607214960819932 0.4897771499597373 0.0896444663382342<br>0.5107037192614156 0.2397520646539955 0.0882962451912425<br>0.5106362379825571 0.7391187967293287 0.0890554573088218<br>0.5127255590650700 0.9912123255701586 0.1820579693198359<br>0.5168731490967391 0.4876795199857571 0.1833384619821517<br>0.7612313254793300 0.9892331297370189 0.0882871458257890<br>0.7592756077984615 0.2377643491227761 0.1820627306504032<br>0.7549395831717027 0.7413210923085584 0.1833169985922803<br>0.7612358704096437 0.4898263744775243 0.0890606407439506<br>0.0029504400716175 0.4814795001563570 0.1857758211968630<br>0.0091795980621652 0.9955015527943635 0.1832857885516624<br>0.1312300653382007 0.6125905316563757 0.3748420306747226<br>0.0335565407522398 0.7149677894616092 0.2839306874676196<br>0.2349376553639130 0.5141716529140681 0.2842570293893051 |

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| STRUCTURE | COORDINATES |
|---|---|
| **$RU_2IR@MGO$** | O Mg Ir Ru<br>16 16 1 2<br>Direct<br>0.0027094774297463 0.0022752453528023 0.0843477856046685<br>0.0028503007201175 0.5007796201560819 0.0855875911991093<br>0.2433303650993853 0.5103633209931697 0.1864203035833934<br>0.2502728140538789 0.9994663047844746 0.1833255787264499<br>0.2543549279705148 0.2533845883253368 0.0898396157792847<br>0.2527813455352028 0.7506398952186086 0.0856125058614550<br>0.5002801293267135 0.4992268674943892 0.0899784554551857<br>0.5016517616439273 0.0028948225902090 0.0861072712503504<br>0.5159752011804795 0.2377459049841921 0.1884064785297328<br>0.5036793271403526 0.7543093638965397 0.1835492699019477<br>0.7544349622112738 0.9991990542172128 0.1833526296550118<br>0.7504883286886310 0.2518656415180032 0.0861230029686269<br>0.7512171046882536 0.7508489114928996 0.0843178847157796<br>0.7542797205531185 0.5034705122812478 0.1833778364223236<br>0.0007801810147100 0.7528447268483743 0.1832402540043054<br>0.9993795440075796 0.2500556106851283 0.1834785955574164<br>0.0027359171523434 0.2519547956509354 0.0884541796353863<br>0.0025366615213159 0.7510784527353549 0.0871593117726159<br>0.2525542832501343 0.0027161655152649 0.0887534157045465<br>0.2501716644094524 0.7578554579393640 0.1819977573298379<br>0.2400584282255711 0.2444391742451486 0.1876305064915062<br>0.2523997478893651 0.5012534323584181 0.0901652717319829<br>0.5017720647981734 0.2518489013572414 0.0900251010105569<br>0.5016885498319514 0.7508540992561138 0.0884840194614444<br>0.5036713128195905 0.9940007163218376 0.1823511061224307<br>0.5093988414487840 0.5137604547993284 0.1876343482252444<br>0.7517796066970223 0.0017982289623689 0.0876886679675040<br>0.7596764191769330 0.2501041745393446 0.1823546098870540<br>0.7545703898443902 0.7534014224626669 0.1811531960058843<br>0.7508905743990719 0.5010915000536738 0.0887928386790343<br>0.9959018763273786 0.5035786739831198 0.1819737077254858<br>0.0003281702566140 0.9991879602587314 0.1811035915830465<br>0.4504433747864768 0.3037474586946658 0.2767625034915626<br>0.3480853861230638 0.4072964958232057 0.3725881021462467<br>0.2503010057784595 0.5036718672045910 0.2879227418136192 |

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|---|---|
| STRUCTURE | Coordinates |
| **$RUIR_2$@MGO** | O Mg Ir Ru<br>16 16 2 1<br>Direct<br>0.0158586736503637 0.0088257569921583 0.0820242679877333<br>0.0161534720960036 0.5074584747419532 0.0837712598946783<br>0.2549608380180116 0.5184435422870943 0.1851447027656566<br>0.2629604133395012 0.0062145713581527 0.1812505048613896<br>0.2671729004269160 0.2606621849256691 0.0883509576352229<br>0.2658925179135698 0.7571805181035566 0.0837977825240569<br>0.5128540474433781 0.5060719248635945 0.0883261757371461<br>0.5144405454847191 0.0102203911500423 0.0840513116373555<br>0.5299170207678686 0.2434688466892357 0.1864824532597673<br>0.5171583488039181 0.7618811943599920 0.1810705471223261<br>0.7675124298650120 0.0057312083195060 0.1809950170310992<br>0.7632072515425593 0.2589463016074530 0.0840120250333465<br>0.7644719352315467 0.7576042982266581 0.0820238152398957<br>0.7671866429330938 0.5105643075991176 0.1811774530393808<br>0.0138837554719871 0.7596083784377420 0.1812111394061937<br>0.0114966462002040 0.2560881288450028 0.1810705054106869<br>0.0155535832421912 0.2585697288132179 0.0863231295486827<br>0.0157042484451628 0.7576834792517930 0.0850388570384544<br>0.2654903416756875 0.0095411665685850 0.0867205098704902<br>0.2635521377163046 0.7659006075056970 0.1801486783642180<br>0.2508186598042433 0.2494637067260487 0.1860827048342693<br>0.2653181017616603 0.5080339778932906 0.0883053360803784<br>0.5144564220652698 0.2588503343001399 0.0881654073278240<br>0.5147197506450083 0.7578312951233924 0.0863418259206459<br>0.5169733388881607 0.0004019317171211 0.1803320178941769<br>0.5238862865414017 0.5226430886512075 0.1860572899552515<br>0.7646828589419293 0.0086098874123404 0.0853670804634392<br>0.7729382586394076 0.2564733044539081 0.1802643459681168<br>0.7679056357557176 0.7605952044320494 0.1789405577079497<br>0.7637909085323923 0.5078792074689866 0.0866584599606705<br>0.0075341803361485 0.5098061234460806 0.1801194381975517<br>0.0127281391463032 0.0054914519685411 0.1789578931144516<br>0.4648137930616284 0.3086344611243099 0.2733468819234455<br>0.3649867072550926 0.4087892846776437 0.3699260200156108<br>0.2642089523576144 0.5096615729587261 0.2823736622284561 |

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| STRUCTURE | Coordinates |
| **$IR_3$@MGO** | O Mg Ir<br>16 16 3<br>Direct<br>0.9871494556190707 0.4932823379247377 0.0892182634940755<br>0.2391637202986816 0.2417403104330101 0.0888759995576665<br>0.9912649352412168 0.2415488049881246 0.1867923812144330<br>0.7395927035004254 0.2456420738016265 0.0891681774473073<br>0.4888831623092295 0.2434739453291432 0.1853054034101961<br>0.2387454756048875 0.7448712120170826 0.0893218814491892<br>0.2355313407283991 0.4906863986102779 0.1858905842434703<br>0.9894327680809775 0.7441870156317195 0.1853062335598627<br>0.7380770933491191 0.7420569854619394 0.0907970921019295<br>0.7514112539765700 0.4815421150325097 0.1944358218166206<br>0.4838282565920673 0.7490816152488380 0.1826777834784047<br>0.4906357604911054 0.4945954551638925 0.0908924292837255<br>0.2330173418580880 -0.0001284120794379 0.1932136125151134<br>0.9911829344094449 0.9935049872852619 0.0887140279497966<br>0.7421510452445904 0.9972883376226088 0.1857205701470501<br>0.4876942814213520 0.9940613449060351 0.0893051053450008<br>0.9854976172740334 0.9946449960697613 0.1853327766035867<br>0.9893197206829404 0.2434911933983726 0.0914752107460365<br>0.2381876339591271 0.2473237528891399 0.1853758097945359<br>0.4894754357855285 0.2440027141204491 0.0916915858600599<br>0.7414469727227172 0.2392688936880391 0.1857442346460726<br>0.9935292708127152 0.4914401569619127 0.1857951634874981<br>0.9888285375655667 0.7432563019395680 0.0916626932279248<br>0.2391210487843581 0.4942495985352485 0.0914598216924783<br>0.2297410955403052 0.7359085294219317 0.1889164497338794<br>0.4790185113860576 0.4855230310494996 0.1893424175474881<br>0.4884743798648605 0.7442680478641678 0.0906582276658892<br>0.7384678365130979 0.4943001519609807 0.0915690783438981<br>0.7472968134832969 0.7537747316494448 0.1893079305066322<br>0.2402122817139944 0.9925116950369217 0.0894632431975282<br>0.4970131935235721 0.0031422121559340 0.1888453934047913<br>0.7385447113067766 0.9936260651563233 0.0913591276190557<br>0.2860308151445360 0.9474021950529190 0.2855618026512720<br>0.4772495116778784 0.7558553786804757 0.2783042238798499<br>0.6657331925334123 0.5670357579915444 0.2804795603776895 |

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**Text S1.** Computation details for computing SOCs, MAE, magnetic moment etc.

We choose the direction perpendicular to the MgO substrate as the z-axis direction and the direction parallel to the substrate as the x-axis direction to create a three-dimensional orthogonal coordinate system. To consider the Coulomb interaction, we use a method of *GGA+U* with *U-J* values for 4d and 5d states (2.42 eV for Ir and 2.34 eV for Ru ) to describe the d electrons[1-3]. We compute SOC in a self-consistent method using the VASP's noncollinear mode. Self-consistent computations with SOC in three directions (x, y and z) were performed in order to achieve MAE. The MAE is defined as the difference between total energy in the perpendicular (z) and in-plane (x) directions considering the SOC. Static calculation to obtain the total spin magnetic moment.

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[3] J. Hu and R. Wu, *Nano Lett.*, 2014, 14, 1853–1858.