Phase transitions and amorphization of M_2AgF_4 (M = Na, K, Rb) compounds at high pressure

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SUPPLEMENTARY MATERIAL

Structures and crystallographic information files for five computationally studied polymorphs of K_2AgF_4

Color legend: gray = Ag, green = F, purple = K). Green lines indicate presence of Ag-F bonds shorter than or equal to 2.2 Å, whereas yellow lines show Ag-F distances between 2.2 and 2.8 Å. Space groups for unit cells were determined with accuracy of 0.05 Å. The unit cell dimensions and volumes of the high-pressure structures are also given as a percent value of the 0 GPa structures.

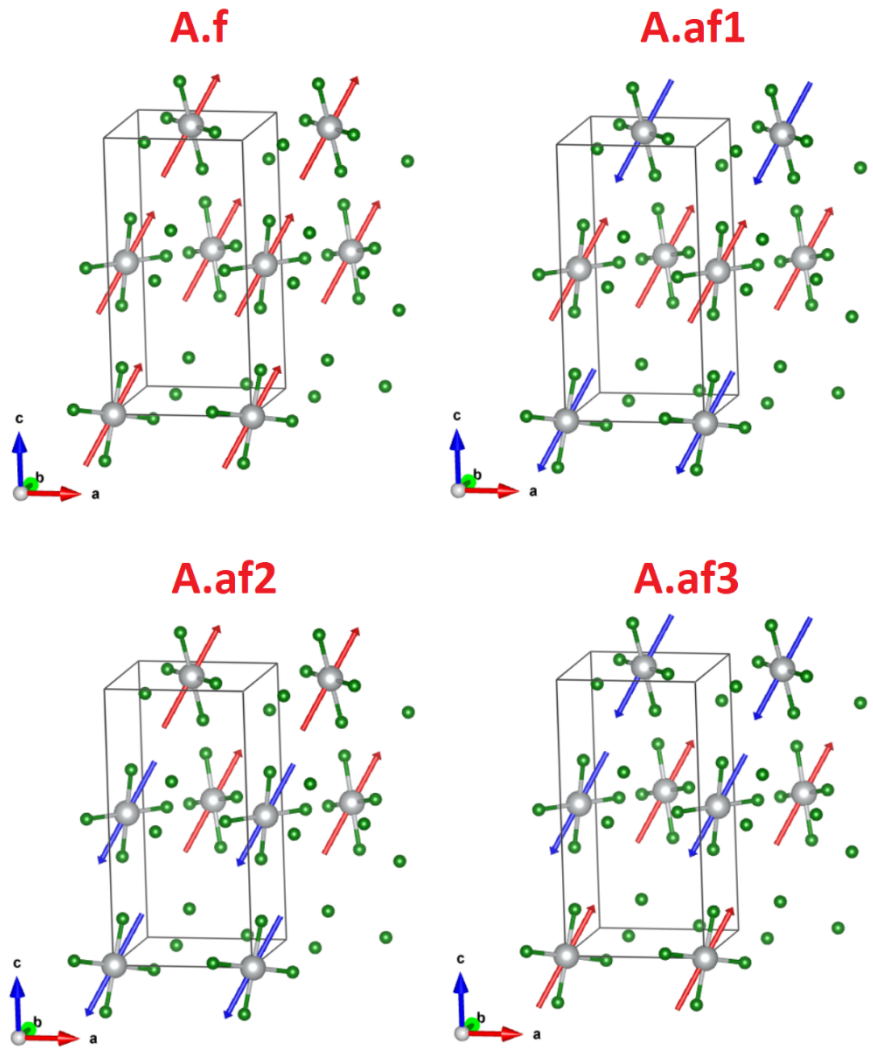

Figure S1. Investigated magnetic variants of K_2AgF_4 **A** structures: ferromagnetic (**A.f**) and antiferromagnetic ones (**A.af1**, **A.af2** and **A.af3**). Only Ag-F sublattices are shown. Ag-F bonds are indicated for interatomic distances not longer than 2.2 Å. All visualized example structures are for p = 0 GPa.

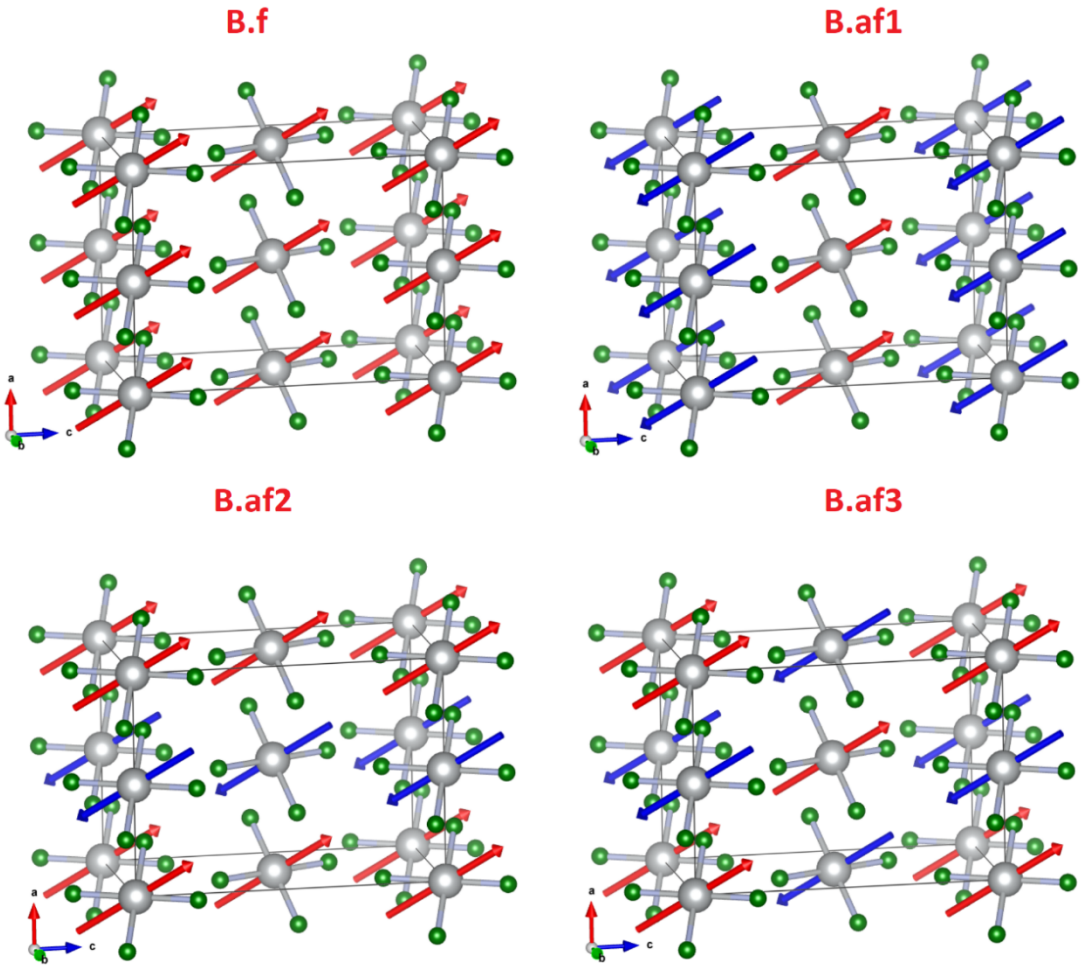

Figure S2. Investigated magnetic variants of K_2AgF_4 **B** structures: ferromagnetic (**B.f**) and antiferromagnetic ones (**B.af1**, **B.af2** and **B.af3**). Only Ag-F sublattices are shown. Ag-F bonds are indicated for interatomic distances not longer than 2.2 Å. All visualized example structures are for p = 0 GPa.

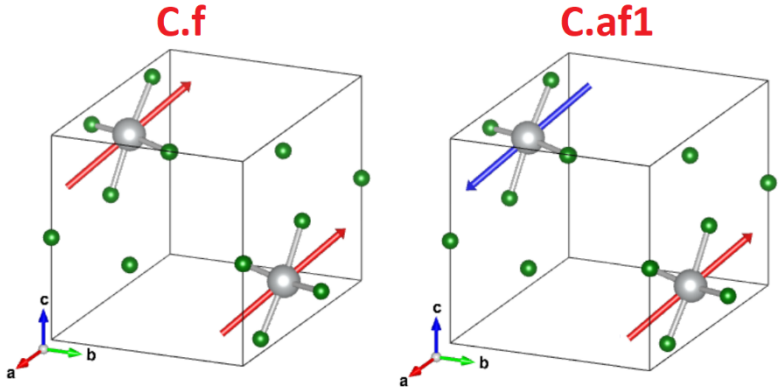

Figure S3. Investigated magnetic variants of K_2AgF_4 **C** structures: ferromagnetic (**C.f**) and antiferromagnetic one (**C.af1**). Only Ag-F sublattices are shown. Ag-F bonds are indicated for interatomic distances not longer than 2.2 Å. All visualized example structures are for p = 0 GPa.

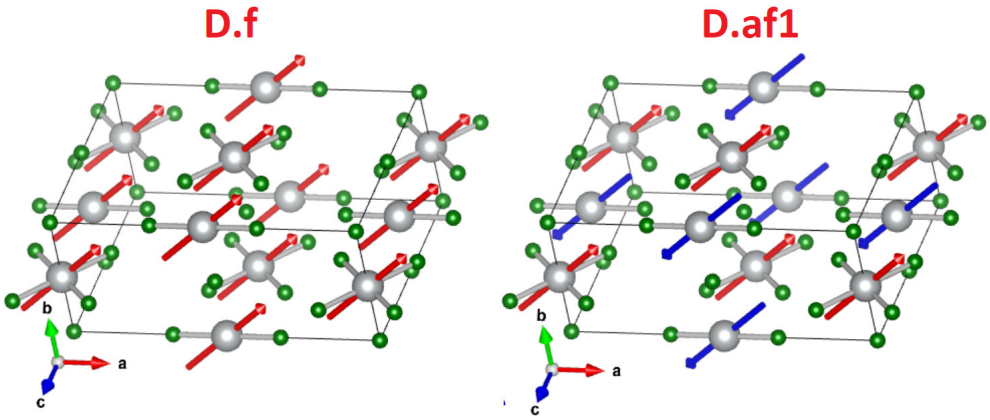

Figure S4. Investigated magnetic variants of K_2AgF_4 **D** structures: ferromagnetic (**D.f**) and antiferromagnetic one (**D.af1**). Only Ag-F sublattices are shown. Ag-F bonds are indicated for interatomic distances not longer than 2.2 Å. All visualized example structures are for p = 0 GPa.

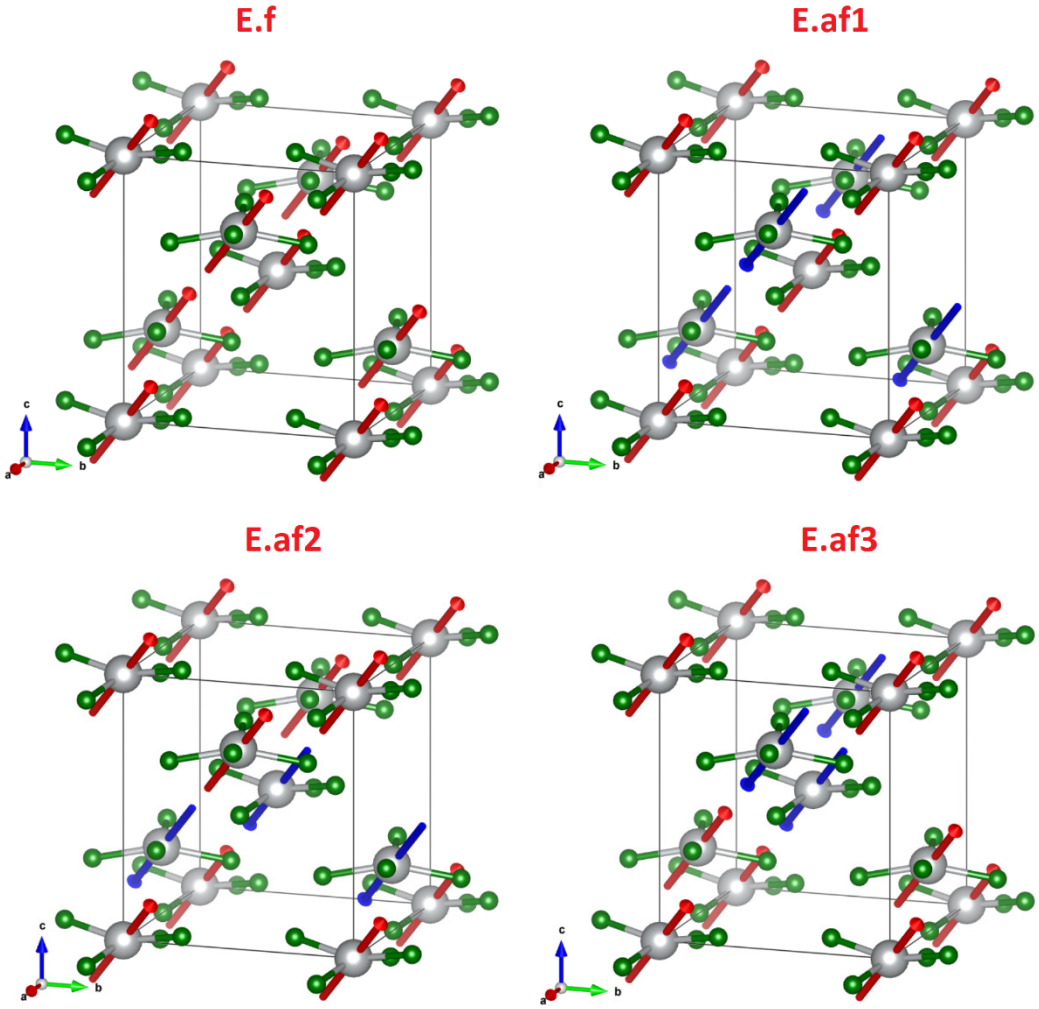

Figure S5. Investigated magnetic variants of K_2AgF_4 **E** structures: ferromagnetic (**E.f**) and antiferromagnetic ones (**E.af1**, **E.af2** and **E.af3**). Only Ag-F sublattices are shown. Ag-F bonds are indicated for interatomic distances not longer than 2.2 Å. All visualized example structures are for p = 0 GPa.

Appendix S1. CIF files of structures discussed in this study.

Ambient pressure structures.

```
## DFT(PBEsol)+U K2AgF4 A p=0GPa magn.: f
_cell_length_a    6.3315500000
_cell_length_b    6.3277300000
_cell_length_c    12.4819800000
_cell_angle_alpha 90.0000000000
_cell_angle_beta  90.0449600000
_cell_angle_gamma 90.0000000000
_cell_volume      500.0821226682

_symmetry_space_group_name_H-M "P 1 21/c 1"
_symmetry_Int_Tables_number 14
_space_group.reference_setting '014:-P 2ybc'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 -x,y+1/2,-z+1/2
3 -x,-y,-z
4 x,-y+1/2,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   2 c 0.00000  0.00000 0.50000 1.00000 0,0,0
Ag2 Ag   2 b 0.50000  0.00000 0.00000 1.00000 0,0,0
F1  F    4 e -0.05844 0.44230 0.83675 1.00000 Dx,Dy,Dz
F2  F    4 e 0.44338  0.55660 0.66343 1.00000 Dx,Dy,Dz
F3  F    4 e 0.77059  0.27159 0.03749 1.00000 Dx,Dy,Dz
F4  F    4 e 0.73305  0.73174 0.50023 1.00000 Dx,Dy,Dz
K1  K    4 e -0.02351 0.47737 0.35968 1.00000 Dx,Dy,Dz
K2  K    4 e 0.52284  0.47663 0.86006 1.00000 Dx,Dy,Dz

## DFT(PBEsol)+U K2AgF4 B p=0GPa magn.: af1
_cell_length_a    3.7142100000
_cell_length_b    10.2182100000
_cell_length_c    6.3768200000
_cell_angle_alpha 90.0000000000
_cell_angle_beta  91.8597600000
_cell_angle_gamma 90.0000000000
_cell_volume      241.8892758234

_symmetry_space_group_name_H-M "P 1 21/c 1"
_symmetry_Int_Tables_number 14
_space_group.reference_setting '014:-P 2ybc'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 -x,y+1/2,-z+1/2
3 -x,-y,-z
4 x,-y+1/2,z+1/2

loop_
```

```
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   2 c 0.00000 0.00000 0.50000 1.00000 0,0,0
F1  F    4 e 0.41352 0.44745 0.21908 1.00000 Dx,Dy,Dz
F2  F    4 e 0.01434 0.68835 0.12556 1.00000 Dx,Dy,Dz
K1  K    4 e 0.51872 0.68186 0.41817 1.00000 Dx,Dy,Dz

## DFT(PBEsol)+U K2AgF4 C p=0GPa magn.: af1
_cell_length_a    6.0329400000
_cell_length_b    6.9289208299
_cell_length_c    7.0826800000
_cell_angle_alpha 94.5327116860
_cell_angle_beta  112.7446700000
_cell_angle_gamma 114.0180519600
_cell_volume      469,79.3826835153

_symmetry_space_group_name_H-M "P -1"
_symmetry_Int_Tables_number 2
_space_group.reference_setting '002:-P 1'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 -x,-y,-z

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   2 i 0.41276 0.04242 0.76334  1.00000 Dx,Dy,Dz
F1  F    2 i 0.62817 0.38244 0.81166  1.00000 Dx,Dy,Dz
F2  F    2 i 0.25256 0.06383 -0.01898 1.00000 Dx,Dy,Dz
F3  F    1 f 0.50000 0.00000 0.50000  1.00000 0,0,0
F4  F    1 b 0.00000 0.00000 0.50000  1.00000 0,0,0
F5  F    2 i 0.81729 0.29566 0.32741  1.00000 Dx,Dy,Dz
K1  K    2 i 0.06630 0.34581 0.76360  1.00000 Dx,Dy,Dz
K2  K    2 i 0.24116 0.24408 0.30540  1.00000 Dx,Dy,Dz

## DFT(PBEsol)+U K2AgF4 D p=0GPa magn.: f
_cell_length_a    11.5964698009
_cell_length_b    4.9061300000
_cell_length_c    8.2557867306
_cell_angle_alpha 90.0000000000
_cell_angle_beta  90.0000000000
_cell_angle_gamma 90.0000000000
_cell_volume      469.7029831963

_symmetry_space_group_name_H-M "I 2/m 2/m 2/m"
_symmetry_Int_Tables_number 71
_space_group.reference_setting '071:-I 2 2'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
```

```
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x,-y,-z
3 -x,y,-z
4 -x,-y,z
5 -x,-y,-z
6 -x,y,z
7 x,-y,z
8 x,y,-z
9 x+1/2,y+1/2,z+1/2
10 x+1/2,-y+1/2,-z+1/2
11 -x+1/2,y+1/2,-z+1/2
12 -x+1/2,-y+1/2,z+1/2
13 -x+1/2,-y+1/2,-z+1/2
14 -x+1/2,y+1/2,z+1/2
15 x+1/2,-y+1/2,z+1/2
16 x+1/2,y+1/2,-z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   2 d 0.50000 0.00000 0.50000 1.00000 0,0,0
Ag2 Ag   2 b 0.00000 0.50000 0.50000 1.00000 0,0,0
F1  F    4 e 0.32453 0.00000 0.00000 1.00000 Dx,0,0
F2  F    8 m 0.62431 0.00000 0.68095 1.00000 Dx,0,Dz
F3  F    2 c 0.50000 0.50000 0.00000 1.00000 0,0,0
F4  F    2 a 0.00000 0.00000 0.00000 1.00000 0,0,0
K1  K    8 m 0.15450 0.00000 0.24926 1.00000 Dx,0,Dz

## DFT(PBEsol)+U K2AgF4 E p=0GPa magn.: f
_cell_length_a    7.2955650000
_cell_length_b    7.2955650000
_cell_length_c    8.2047100000
_cell_angle_alpha 90.0000000000
_cell_angle_beta  90.0000000000
_cell_angle_gamma 90.0000000000
_cell_volume      436.6978941031

_symmetry_space_group_name_H-M "I -4 2 d"
_symmetry_Int_Tables_number 122
_space_group.reference_setting '122:I -4 2bw'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x,-y+1/2,-z+1/4
3 -x,y+1/2,-z+1/4
4 -x,-y,z
5 y,x+1/2,z+1/4
6 y,-x,-z
7 -y,x,-z
8 -y,-x+1/2,z+1/4
9 x+1/2,y+1/2,z+1/2
10 x+1/2,-y,-z+3/4
11 -x+1/2,y,-z+3/4
12 -x+1/2,-y+1/2,z+1/2
13 y+1/2,x,z+3/4
14 y+1/2,-x+1/2,-z+1/2
```

```
15 -y+1/2,x+1/2,-z+1/2
16 -y+1/2,-x,z+3/4

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   4 b 0.00000  0.00000 0.50000 1.00000 0,0,0
F1  F   16 e -0.06852 0.27709 0.45335 1.00000 Dx,Dy,Dz
K1  K    8 d 0.88647  0.25000 0.12500 1.00000 Dx,0,0
```

Relevant elevated pressure structures.

```
## DFT(PBEsol)+U K2AgF4 E p=10GPa magn.: f
_cell_length_a    6.9150650000
_cell_length_b    6.9150650000
_cell_length_c    7.6345600000
_cell_angle_alpha 90.0000000000
_cell_angle_beta  90.0000000000
_cell_angle_gamma 90.0000000000
_cell_volume      365.0703364160

_symmetry_space_group_name_H-M "I -4 2 d"
_symmetry_Int_Tables_number 122
_space_group.reference_setting '122:I -4 2bw'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x,-y+1/2,-z+1/4
3 -x,y+1/2,-z+1/4
4 -x,-y,z
5 y,x+1/2,z+1/4
6 y,-x,-z
7 -y,x,-z
8 -y,-x+1/2,z+1/4
9 x+1/2,y+1/2,z+1/2
10 x+1/2,-y,-z+3/4
11 -x+1/2,y,-z+3/4
12 -x+1/2,-y+1/2,z+1/2
13 y+1/2,x,z+3/4
14 y+1/2,-x+1/2,-z+1/2
15 -y+1/2,x+1/2,-z+1/2
16 -y+1/2,-x,z+3/4

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   4 b 0.00000  0.00000 0.50000 1.00000 0,0,0
F1  F   16 e -0.07345 0.29072 0.45910 1.00000 Dx,Dy,Dz
K1  K    8 d 0.89743  0.25000 0.12500 1.00000 Dx,0,0
```

```
## DFT(PBEsol)+U K2AgF4 C p=60GPa magn.: f
_cell_length_a    4.6368700000
_cell_length_b    5.9522700000
_cell_length_c    5.9543300000
_cell_angle_alpha 113.5047100000
_cell_angle_beta  103.7115500000
_cell_angle_gamma 105.6390100000
_cell_volume      133.6222366842

_symmetry_space_group_name_H-M "P -1"
_symmetry_Int_Tables_number 2
_space_group.reference_setting '002:-P 1'
_space_group.transform_Pp_abc a,b,c;0,0,0

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 -x,-y,-z

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
_atom_site_fract_symmform
Ag1 Ag   2 i -0.05026 0.85790  0.22323 1.00000 Dx,Dy,Dz
F1  F    2 i 0.76638  0.82685  0.85014 1.00000 Dx,Dy,Dz
F2  F    2 i 0.54524  -0.04823 0.30126 1.00000 Dx,Dy,Dz
F3  F    1 c 0.00000  0.50000  0.00000 1.00000 0,0,0
F4  F    1 e 0.50000  0.50000  0.00000 1.00000 0,0,0
F5  F    2 i 0.08766  0.29885  0.53133 1.00000 Dx,Dy,Dz
K1  K    2 i 0.25976  0.75140  0.85265 1.00000 Dx,Dy,Dz
K2  K    2 i 0.62187  0.37185  0.59830 1.00000 Dx,Dy,Dz
```