Electronic Supplementary Information for:

The first molecular superconductor based on BEDT-TTF radical cation salt with paramagnetic tris(oxalato)ruthenate anion

Tatiana G. Prokhorova,[a] Leokadiya V. Zorina,[b,c] Sergey V. Simonov,[b,c] Vladimir N. Zverev,[b,c] Enric Canadell,[d] Rimma P. Shibaeva,[b] Eduard B. Yagubskii[a]

[d] *Institute of Problems of Chemical Physics, RAS, Chernogolovka, MD,142432, Russia*
[b] *Institute of Solid State Physics, RAS, Chernogolovka, MD, 142432, Russia*
[c] *Moscow Institute of Physics and Technology, Dolgoprudny, Moscow region, Russia*
[d] *Institut de Ciència de Materials de Barcelona, CSIC, Campus de la UAB, E-08193 Bellaterra, Spain*

The X-ray single crystal diffraction study and electronic band structure calculations have also been carried out for the sample #4 of the compound **1** with minimal T_c = 2.8 K to compare with those for the sample #1 showing maximal T_c = 6.3 K and described in the article (see Fig. 8 for difference in temperature dependence of resistivity). The crystal structure data for the sample #4 are reported below as a crystallographic information file; calculated transfer integrals, band structure and Fermi surface are reported in Table S1 and Figures S3-S4. The results for the sample #4 are practically undistinguishable from ones obtained for the sample #1 so that we must conclude that there is nothing in the crystal and electronic structure of the samples justifying the observed differences in physical behavior.

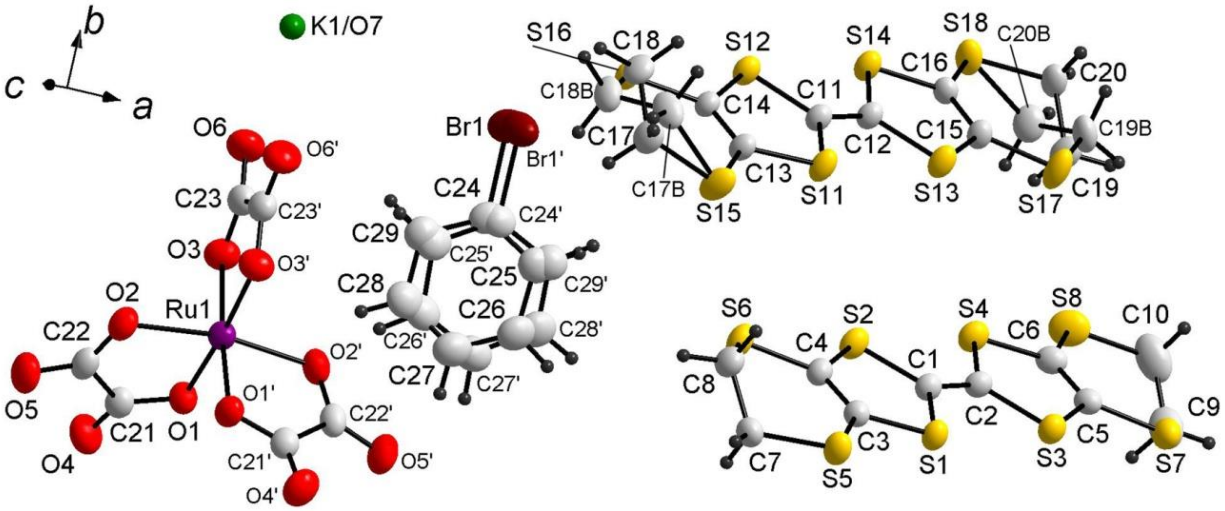

Figure S1. Asymmetric unit of the structure **1**, sample #1. Displacement ellipsoids are drawn at the 50% probability level. The same numbering scheme is used for the structure **1**, sample #4.

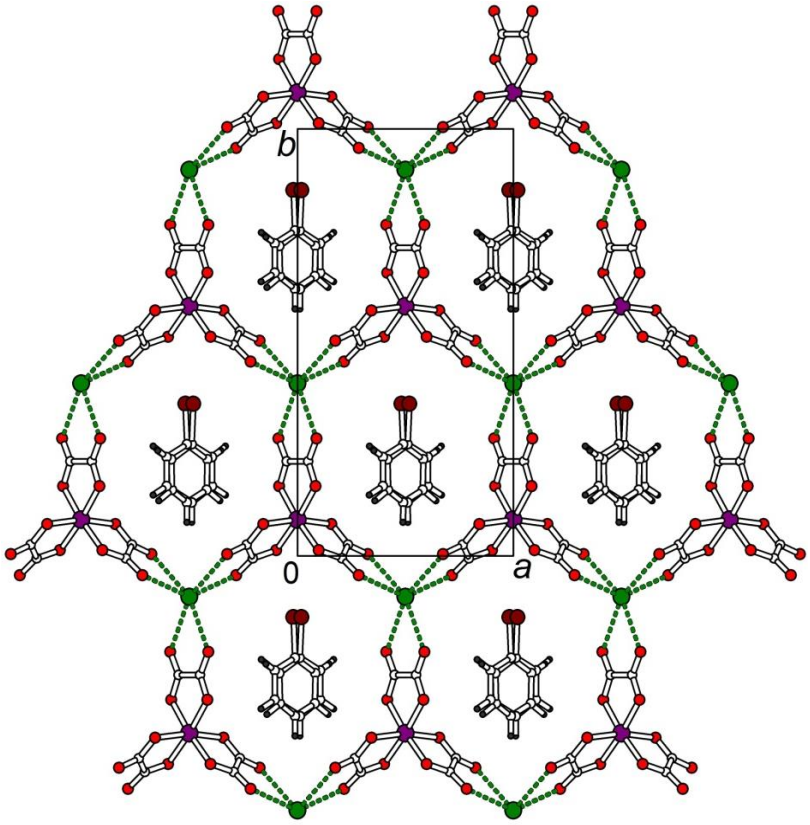

Fig. S2. Honeycomb-like anion layer in the structure **1**.

Crystal structure (CIF-file) of β''-(BEDT-TTF)$_4$K$_x$(H$_3$O)$_{1-x}$[Ru^{III}(ox)$_3$]C$_6$H$_5$Br (**1**, x = 0.77, sample #4 with minimal T_c):

```
data_Ru(ox)3-sample4-RT

_audit_creation_method           SHELXL-97
_chemical_name_systematic        ?
_chemical_name_common            ?
_chemical_melting_point          ?
_chemical_formula_moiety
'4(C10 H8 S8), C6 Ru O12, C6 H5 Br, K0.77, 0.23(H3 O)'
_chemical_formula_sum
 'C52 H37.69 Br K0.77 O12.23 Ru S32'
_chemical_formula_weight         2095.20

loop_
 _atom_type_symbol
 _atom_type_description
 _atom_type_scat_dispersion_real
 _atom_type_scat_dispersion_imag
 _atom_type_scat_source
'C'  'C'   0.0033   0.0016
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'H'  'H'   0.0000   0.0000
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'O'  'O'   0.0106   0.0060
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'Ru'  'Ru'  -1.2594   0.8363
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'S'  'S'   0.1246   0.1234
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'Br'  'Br'  -0.2901   2.4595
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'
'K'  'K'   0.2009   0.2494
'International Tables Vol C Tables 4.2.6.8 and 6.1.1.4'

_symmetry_cell_setting           monoclinic
_symmetry_space_group_name_H-M    'C 2/c'
_symmetry_space_group_name_Hall   '-C 2yc'

loop_
 _symmetry_equiv_pos_as_xyz
 'x, y, z'
 '-x, y, -z+1/2'
 'x+1/2, y+1/2, z'
 '-x+1/2, y+1/2, -z+1/2'
 '-x, -y, -z'
 'x, -y, z-1/2'
 '-x+1/2, -y+1/2, -z'
```

```
 'x+1/2, -y+1/2, z-1/2'

_cell_length_a                 10.2826(7)
_cell_length_b                 19.9221(16)
_cell_length_c                 35.555(3)
_cell_angle_alpha               90.00
_cell_angle_beta                93.271(6)
_cell_angle_gamma               90.00
_cell_volume                   7271.6(9)
_cell_formula_units_Z           4
_cell_measurement_temperature     293(2)
_cell_measurement_reflns_used     10700
_cell_measurement_theta_min       1.9620
_cell_measurement_theta_max       32.2270

_exptl_crystal_description       rhomb
_exptl_crystal_colour            black
_exptl_crystal_size_max          0.33
_exptl_crystal_size_mid          0.28
_exptl_crystal_size_min          0.16
_exptl_crystal_density_meas      ?
_exptl_crystal_density_diffrn    1.914
_exptl_crystal_density_method    'not measured'
_exptl_crystal_F_000             4213
_exptl_absorpt_coefficient_mu    1.787
_exptl_absorpt_correction_type   'multi-scan'
_exptl_absorpt_correction_T_min  0.50104
_exptl_absorpt_correction_T_max  1.00000
_exptl_absorpt_process_details
;
CrysAlisPro, Agilent Technologies,
Version 1.171.36.24 (release 03-12-2012 CrysAlis171 .NET)
(compiled Dec  3 2012,18:21:49)
Empirical absorption correction using spherical harmonics,
 implemented in SCALE3 ABSPACK scaling algorithm.
;

_exptl_special_details
;
 ?
;

_diffrn_ambient_temperature      293(2)
_diffrn_radiation_wavelength     0.71073
_diffrn_radiation_type           MoK\a
_diffrn_radiation_source         'Enhance (Mo) X-ray Source'
_diffrn_radiation_monochromator  graphite
_diffrn_measurement_device_type  'Xcalibur, Ruby, Gemini'
_diffrn_measurement_method       ?
_diffrn_detector_area_resol_mean  10.4752
_diffrn_standards_number         ?
_diffrn_standards_interval_count  ?
```

```
_diffrn_standards_interval_time   ?
_diffrn_standards_decay_%         ?
_diffrn_reflns_number             29544
_diffrn_reflns_av_R_equivalents   0.0346
_diffrn_reflns_av_sigmaI/netI     0.0415
_diffrn_reflns_limit_h_min        -14
_diffrn_reflns_limit_h_max        14
_diffrn_reflns_limit_k_min        -29
_diffrn_reflns_limit_k_max        30
_diffrn_reflns_limit_l_min        -32
_diffrn_reflns_limit_l_max        51
_diffrn_reflns_theta_min          2.04
_diffrn_reflns_theta_max          32.94
_diffrn_measured_fraction_theta_max    0.900
_diffrn_reflns_theta_full         26.00
_diffrn_measured_fraction_theta_full   0.995
_reflns_number_total              12270
_reflns_number_gt                 9248
_reflns_threshold_expression      >2\s(I)

_computing_data_collection        'CrysAlis CCD, Oxford Diffraction Ltd.'
_computing_cell_refinement        'CrysAlis RED, Oxford Diffraction Ltd.'
_computing_data_reduction         'CrysAlis RED, Oxford Diffraction Ltd.'
_computing_structure_solution     'SHELXS-97 (Sheldrick, 2008)'
_computing_structure_refinement   'SHELXL-97 (Sheldrick, 2008)'
_computing_molecular_graphics
'Ortep-3 for Windows (Farrugia, 1997), DIAMOND 3.1 (Crystal Impact GbR 2005)'
_computing_publication_material   'WinGX publication routines (Farrugia, 1999)'

_refine_special_details
;
 Refinement of F^2^ against ALL reflections.  The weighted R-factor wR and
 goodness of fit S are based on F^2^, conventional R-factors R are based
 on F, with F set to zero for negative F^2^. The threshold expression of
 F^2^ > 2\s(F^2^) is used only for calculating R-factors(gt) etc. and is
 not relevant to the choice of reflections for refinement.  R-factors based
 on F^2^ are statistically about twice as large as those based on F, and R-
 factors based on ALL data will be even larger.
;

_refine_ls_structure_factor_coef  Fsqd
_refine_ls_matrix_type            full
_refine_ls_weighting_scheme       calc
_refine_ls_weighting_details
 'calc w=1/[\s^2^(Fo^2^)+(0.0570P)^2^+10.8807P] where P=(Fo^2^+2Fc^2^)/3'
_atom_sites_solution_primary      direct
_atom_sites_solution_secondary    difmap
_atom_sites_solution_hydrogens    geom
_refine_ls_hydrogen_treatment     constr
_refine_ls_extinction_method      none
_refine_ls_extinction_coef        ?
_refine_ls_number_reflns          12270
```

```
_refine_ls_number_parameters      518
_refine_ls_number_restraints      86
_refine_ls_R_factor_all           0.0725
_refine_ls_R_factor_gt            0.0509
_refine_ls_wR_factor_ref          0.1295
_refine_ls_wR_factor_gt           0.1182
_refine_ls_goodness_of_fit_ref    1.013
_refine_ls_restrained_S_all       1.020
_refine_ls_shift/su_max           0.014
_refine_ls_shift/su_mean          0.001
_refine_diff_density_max          1.820
_refine_diff_density_min          -1.718
_refine_diff_density_rms          0.101

loop_
 _atom_site_label
 _atom_site_type_symbol
 _atom_site_fract_x
 _atom_site_fract_y
 _atom_site_fract_z
 _atom_site_U_iso_or_equiv
 _atom_site_adp_type
 _atom_site_occupancy
 _atom_site_symmetry_multiplicity
 _atom_site_calc_flag
 _atom_site_refinement_flags
 _atom_site_disorder_assembly
 _atom_site_disorder_group
S1 S 1.16594(6) 0.14344(4) 0.031621(19) 0.03327(15) Uani 1 1 d . . .
S2 S 0.92304(7) 0.21384(4) 0.00908(2) 0.03662(16) Uani 1 1 d . . .
S3 S 1.27731(6) 0.17359(4) -0.04914(2) 0.03467(15) Uani 1 1 d . . .
S4 S 1.03692(7) 0.24508(4) -0.07270(2) 0.03668(16) Uani 1 1 d . . .
S5 S 1.09277(7) 0.11074(4) 0.10863(2) 0.03989(17) Uani 1 1 d . . .
S6 S 0.79915(7) 0.19278(4) 0.08039(2) 0.04294(18) Uani 1 1 d . . .
S7 S 1.40273(8) 0.19241(4) -0.12075(2) 0.04638(19) Uani 1 1 d . . .
S8 S 1.10938(10) 0.27238(5) -0.14980(2) 0.0546(2) Uani 1 1 d . . .
C1 C 1.0763(2) 0.18751(13) -0.00324(7) 0.0288(5) Uani 1 1 d . . .
C2 C 1.1241(2) 0.20078(13) -0.03743(7) 0.0287(5) Uani 1 1 d . . .
C3 C 1.0477(2) 0.14755(13) 0.06505(7) 0.0289(5) Uani 1 1 d . . .
C4 C 0.9363(2) 0.17990(14) 0.05458(8) 0.0305(5) Uani 1 1 d . . .
C5 C 1.2672(3) 0.20708(14) -0.09455(8) 0.0321(5) Uani 1 1 d . . .
C6 C 1.1563(3) 0.23894(14) -0.10565(7) 0.0338(6) Uani 1 1 d . . .
C7 C 0.9393(3) 0.10681(15) 0.13027(9) 0.0386(6) Uani 1 1 d . . .
H7A H 0.8874 0.0712 0.1184 0.046 Uiso 1 1 calc R . .
H7B H 0.9553 0.0949 0.1566 0.046 Uiso 1 1 calc R . .
C8 C 0.8614(3) 0.17131(16) 0.12793(9) 0.0416(7) Uani 1 1 d . . .
H8A H 0.9160 0.2078 0.1377 0.050 Uiso 1 1 calc R . .
H8B H 0.7885 0.1673 0.1439 0.050 Uiso 1 1 calc R . .
C9 C 1.3254(4) 0.1964(2) -0.16742(10) 0.0623(11) Uani 1 1 d . . .
H9A H 1.3910 0.1906 -0.1857 0.075 Uiso 1 1 calc R . .
H9B H 1.2640 0.1595 -0.1706 0.075 Uiso 1 1 calc R . .
C10 C 1.2551(4) 0.2610(2) -0.17522(10) 0.0671(12) Uani 1 1 d . . .
```

H10A H 1.2317 0.2635 -0.2020 0.081 Uiso 1 1 calc R . .
H10B H 1.3143 0.2977 -0.1690 0.081 Uiso 1 1 calc R . .
S11 S 0.85559(6) 0.38312(4) 0.060223(18) 0.03509(15) Uani 1 1 d . . .
S12 S 0.61807(6) 0.45765(4) 0.036588(19) 0.03282(14) Uani 1 1 d . . .
S13 S 0.97306(6) 0.41155(4) -0.019785(19) 0.03546(16) Uani 1 1 d . . .
S14 S 0.73822(6) 0.48751(4) -0.044167(19) 0.03370(15) Uani 1 1 d . . .
S15 S 0.76381(9) 0.34335(5) 0.13452(2) 0.0485(2) Uani 1 1 d . . .
S16 S 0.48507(7) 0.42835(4) 0.10651(2) 0.03845(16) Uani 1 1 d . . .
S17 S 1.10347(8) 0.42838(5) -0.09008(2) 0.0518(2) Uani 1 1 d . . .
S18 S 0.82193(7) 0.51649(4) -0.12088(2) 0.04243(19) Uani 1 1 d . . .
C11 C 0.7693(2) 0.42775(13) 0.02485(7) 0.0279(5) Uani 1 1 d . . .
C12 C 0.8196(2) 0.44038(14) -0.00906(7) 0.0281(5) Uani 1 1 d . . .
C13 C 0.7305(3) 0.38337(13) 0.09155(7) 0.0303(5) Uani 1 1 d . A .
C14 C 0.6209(2) 0.41711(13) 0.08020(7) 0.0283(5) Uani 1 1 d . A .
C15 C 0.9663(2) 0.44496(14) -0.06518(7) 0.0302(5) Uani 1 1 d . B .
C16 C 0.8585(2) 0.48013(13) -0.07671(7) 0.0284(5) Uani 1 1 d . B .
C17 C 0.6497(6) 0.3747(3) 0.16538(15) 0.0413(11) Uani 0.68 1 d PD A 1
H17A H 0.5859 0.3398 0.1692 0.050 Uiso 0.68 1 calc PR A 1
H17B H 0.6950 0.3835 0.1896 0.050 Uiso 0.68 1 calc PR A 1
C18 C 0.5779(6) 0.4376(3) 0.15292(13) 0.0414(10) Uani 0.68 1 d PD A 1
H18A H 0.6401 0.4739 0.1512 0.050 Uiso 0.68 1 calc PR A 1
H18B H 0.5178 0.4498 0.1719 0.050 Uiso 0.68 1 calc PR A 1
C19 C 1.0377(7) 0.4442(3) -0.1381(2) 0.0443(15) Uani 0.68 1 d PD B 1
H19A H 1.1080 0.4414 -0.1552 0.053 Uiso 0.68 1 calc PR B 1
H19B H 0.9750 0.4094 -0.1452 0.053 Uiso 0.68 1 calc PR B 1
C20 C 0.9725(4) 0.5117(3) -0.14283(13) 0.0390(10) Uani 0.68 1 d PD B 1
H20A H 0.9575 0.5211 -0.1695 0.047 Uiso 0.68 1 calc PR B 1
H20B H 1.0307 0.5460 -0.1322 0.047 Uiso 0.68 1 calc PR B 1
C17B C 0.6716(11) 0.4061(7) 0.1620(3) 0.042(3) Uani 0.32 1 d PD A 2
H17C H 0.6861 0.3966 0.1887 0.050 Uiso 0.32 1 calc PR A 2
H17D H 0.7058 0.4506 0.1575 0.050 Uiso 0.32 1 calc PR A 2
C18B C 0.5259(10) 0.4060(6) 0.1520(3) 0.041(2) Uani 0.32 1 d PD A 2
H18C H 0.4840 0.4367 0.1686 0.049 Uiso 0.32 1 calc PR A 2
H18D H 0.4921 0.3614 0.1564 0.049 Uiso 0.32 1 calc PR A 2
C19B C 1.0748(12) 0.4677(8) -0.1326(4) 0.042(3) Uani 0.32 1 d PD B 2
H19C H 1.0995 0.5145 -0.1300 0.050 Uiso 0.32 1 calc PR B 2
H19D H 1.1295 0.4475 -0.1509 0.050 Uiso 0.32 1 calc PR B 2
C20B C 0.9346(10) 0.4637(6) -0.1474(3) 0.046(2) Uani 0.32 1 d PD B 2
H20C H 0.9062 0.4173 -0.1465 0.055 Uiso 0.32 1 calc PR B 2
H20D H 0.9291 0.4775 -0.1736 0.055 Uiso 0.32 1 calc PR B 2
Ru1 Ru 0.0000 0.084225(18) 0.2500 0.03684(9) Uani 1 2 d S . .
K1 K 0.0000 0.40381(6) 0.2500 0.0496(3) Uani 0.77 2 d SP . 1
O7 O 0.0000 0.40381(6) 0.2500 0.0496(3) Uani 0.23 2 d SP C 2
C21 C -0.2062(3) -0.00631(17) 0.23253(8) 0.0398(6) Uani 1 1 d . . .
C22 C 0.2343(3) 0.02580(17) 0.22890(9) 0.0421(7) Uani 1 1 d . . .
C23 C -0.0501(3) 0.22105(18) 0.23236(10) 0.0474(7) Uani 1 1 d . . .
O1 O -0.0978(2) 0.01268(12) 0.21920(6) 0.0436(5) Uani 1 1 d . . .
O2 O 0.1579(2) 0.07427(12) 0.21776(6) 0.0462(5) Uani 1 1 d . . .
O3 O -0.0838(2) 0.16273(13) 0.22024(6) 0.0499(6) Uani 1 1 d . . .
O4 O -0.2833(2) -0.04537(14) 0.21744(7) 0.0572(6) Uani 1 1 d . . .
O5 O 0.3256(3) 0.00370(16) 0.21188(8) 0.0629(7) Uani 1 1 d . . .
O6 O -0.0889(3) 0.27458(14) 0.21912(9) 0.0662(7) Uani 1 1 d . . .

Br1 Br 0.4809(4) 0.35558(4) 0.24655(15) 0.0686(8) Uani 0.50 1 d PU . .
C24 C 0.4859(19) 0.2608(3) 0.2501(10) 0.047(2) Uani 0.50 1 d PU . .
C25 C 0.5844(17) 0.2245(11) 0.2328(6) 0.051(3) Uani 0.50 1 d PU . .
H25 H 0.6481 0.2458 0.2195 0.061 Uiso 0.50 1 calc PR . .
C26 C 0.5804(12) 0.1538(7) 0.2367(3) 0.057(2) Uani 0.50 1 d PU . .
H26 H 0.6407 0.1272 0.2251 0.069 Uiso 0.50 1 calc PR . .
C27 C 0.4888(14) 0.1251(4) 0.2574(3) 0.059(2) Uani 0.50 1 d PU . .
H27 H 0.4882 0.0787 0.2602 0.071 Uiso 0.50 1 calc PR . .
C28 C 0.3949(13) 0.1636(7) 0.2746(3) 0.060(3) Uani 0.50 1 d PU . .
H28 H 0.3329 0.1426 0.2886 0.072 Uiso 0.50 1 calc PR . .
C29 C 0.394(2) 0.2316(12) 0.2709(7) 0.054(3) Uani 0.50 1 d PU . .
H29 H 0.3327 0.2575 0.2824 0.065 Uiso 0.50 1 calc PRU . .

loop_
 _atom_site_aniso_label
 _atom_site_aniso_U_11
 _atom_site_aniso_U_22
 _atom_site_aniso_U_33
 _atom_site_aniso_U_23
 _atom_site_aniso_U_13
 _atom_site_aniso_U_12
S1 0.0268(3) 0.0432(4) 0.0304(3) 0.0073(3) 0.0064(2) 0.0073(3)
S2 0.0293(3) 0.0469(4) 0.0338(3) 0.0030(3) 0.0031(2) 0.0099(3)
S3 0.0285(3) 0.0415(4) 0.0343(3) 0.0061(3) 0.0041(2) 0.0066(3)
S4 0.0326(3) 0.0445(4) 0.0327(3) 0.0054(3) 0.0001(3) 0.0096(3)
S5 0.0336(3) 0.0513(4) 0.0355(4) 0.0127(3) 0.0093(3) 0.0078(3)
S6 0.0302(3) 0.0552(5) 0.0446(4) 0.0010(3) 0.0118(3) 0.0103(3)
S7 0.0465(4) 0.0439(4) 0.0511(5) 0.0029(3) 0.0229(4) 0.0018(3)
S8 0.0742(6) 0.0599(5) 0.0289(4) 0.0093(3) -0.0040(4) 0.0083(5)
C1 0.0253(11) 0.0334(13) 0.0277(12) -0.0014(10) 0.0013(9) 0.0026(10)
C2 0.0275(11) 0.0317(13) 0.0268(12) 0.0006(9) 0.0002(9) 0.0032(10)
C3 0.0296(12) 0.0304(12) 0.0272(12) 0.0021(9) 0.0059(9) -0.0002(10)
C4 0.0275(11) 0.0312(13) 0.0333(13) -0.0028(10) 0.0049(10) 0.0016(10)
C5 0.0350(13) 0.0291(13) 0.0329(13) 0.0009(10) 0.0085(10) -0.0008(10)
C6 0.0427(15) 0.0329(14) 0.0258(12) 0.0007(10) 0.0010(10) -0.0004(11)
C7 0.0380(14) 0.0379(15) 0.0415(16) 0.0055(12) 0.0157(12) -0.0022(12)
C8 0.0412(15) 0.0450(17) 0.0402(16) -0.0014(13) 0.0154(12) 0.0050(13)
C9 0.069(2) 0.074(3) 0.046(2) -0.0183(18) 0.0204(17) -0.017(2)
C10 0.077(3) 0.092(3) 0.0321(17) 0.0145(18) 0.0010(17) -0.029(2)
S11 0.0291(3) 0.0520(4) 0.0248(3) 0.0060(3) 0.0074(2) 0.0140(3)
S12 0.0286(3) 0.0405(4) 0.0299(3) 0.0048(3) 0.0063(2) 0.0098(3)
S13 0.0299(3) 0.0515(4) 0.0255(3) 0.0075(3) 0.0055(2) 0.0132(3)
S14 0.0286(3) 0.0444(4) 0.0287(3) 0.0073(3) 0.0068(2) 0.0111(3)
S15 0.0613(5) 0.0551(5) 0.0308(4) 0.0133(3) 0.0167(3) 0.0246(4)
S16 0.0313(3) 0.0410(4) 0.0448(4) 0.0015(3) 0.0174(3) 0.0063(3)
S17 0.0364(4) 0.0814(6) 0.0392(4) 0.0194(4) 0.0168(3) 0.0256(4)
S18 0.0310(3) 0.0570(5) 0.0400(4) 0.0243(3) 0.0081(3) 0.0065(3)
C11 0.0278(11) 0.0340(13) 0.0220(11) 0.0001(9) 0.0033(9) 0.0048(10)
C12 0.0270(11) 0.0357(13) 0.0219(11) 0.0016(9) 0.0045(9) 0.0046(10)
C13 0.0327(12) 0.0325(13) 0.0267(12) 0.0036(10) 0.0111(10) 0.0076(10)
C14 0.0275(11) 0.0306(12) 0.0275(12) 0.0008(9) 0.0085(9) 0.0029(10)
C15 0.0269(11) 0.0369(14) 0.0275(12) 0.0054(10) 0.0065(9) 0.0058(10)

C16 0.0266(11) 0.0318(13) 0.0273(12) 0.0074(9) 0.0052(9) 0.0017(10)
C17 0.047(3) 0.050(3) 0.028(2) 0.003(2) 0.0120(19) 0.007(3)
C18 0.052(3) 0.042(3) 0.031(2) -0.0106(19) 0.012(2) 0.004(2)
C19 0.048(4) 0.051(4) 0.035(3) 0.006(3) 0.012(3) 0.016(3)
C20 0.034(2) 0.048(3) 0.036(2) 0.0183(19) 0.0125(16) 0.0057(19)
C17B 0.052(6) 0.044(6) 0.030(5) -0.012(5) 0.007(4) -0.009(6)
C18B 0.048(6) 0.047(6) 0.030(5) -0.007(4) 0.012(4) -0.001(5)
C19B 0.040(7) 0.052(8) 0.035(7) 0.025(6) 0.021(5) 0.008(5)
C20B 0.054(6) 0.055(6) 0.029(4) 0.001(4) 0.008(4) -0.008(5)
Ru1 0.04214(18) 0.04251(19) 0.02658(15) 0.000 0.00803(12) 0.000
K1 0.0460(6) 0.0406(6) 0.0641(8) 0.000 0.0197(6) 0.000
O7 0.0460(6) 0.0406(6) 0.0641(8) 0.000 0.0197(6) 0.000
C21 0.0383(15) 0.0467(17) 0.0348(15) 0.0060(12) 0.0050(11) 0.0051(13)
C22 0.0395(15) 0.0532(19) 0.0345(15) -0.0088(13) 0.0091(12) -0.0110(14)
C23 0.0509(18) 0.0478(18) 0.0455(18) 0.0020(14) 0.0188(14) -0.0006(15)
O1 0.0447(12) 0.0536(13) 0.0338(11) -0.0073(9) 0.0135(9) -0.0040(10)
O2 0.0468(12) 0.0582(14) 0.0347(11) 0.0042(10) 0.0128(9) -0.0087(11)
O3 0.0607(14) 0.0511(14) 0.0379(12) 0.0046(10) 0.0026(10) 0.0058(11)
O4 0.0485(13) 0.0693(17) 0.0538(15) -0.0031(13) 0.0035(11) -0.0144(13)
O5 0.0574(15) 0.0810(19) 0.0533(15) -0.0096(14) 0.0294(12) -0.0017(14)
O6 0.0721(18) 0.0545(16) 0.0732(19) 0.0162(14) 0.0128(14) 0.0085(14)
Br1 0.096(2) 0.0543(4) 0.0587(16) 0.0024(6) 0.0345(14) 0.0075(6)
C24 0.049(7) 0.049(3) 0.042(2) 0.001(5) -0.008(6) 0.007(4)
C25 0.045(5) 0.056(5) 0.049(5) 0.000(4) -0.010(4) -0.002(4)
C26 0.048(4) 0.060(5) 0.063(5) 0.002(4) -0.006(4) 0.009(4)
C27 0.059(5) 0.054(3) 0.064(6) 0.001(3) -0.005(5) 0.005(4)
C28 0.058(5) 0.062(5) 0.058(5) -0.002(4) -0.007(4) -0.007(4)
C29 0.056(5) 0.058(5) 0.048(4) 0.000(4) -0.006(4) 0.000(4)

_geom_special_details
;
All s.u.'s (except the s.u. in the dihedral angle between two l.s. planes)
are estimated using the full covariance matrix. The cell s.u.'s are taken
into account individually in the estimation of s.u.'s in distances, angles
and torsion angles; correlations between s.u.'s in cell parameters are only
used when they are defined by crystal symmetry. An approximate (isotropic)
treatment of cell s.u.'s is used for estimating s.u.'s involving l.s. planes.
;

loop_
_geom_bond_atom_site_label_1
_geom_bond_atom_site_label_2
_geom_bond_distance
_geom_bond_site_symmetry_2
_geom_bond_publ_flag
S1 C1 1.740(3) . ?
S1 C3 1.750(3) . ?
S2 C1 1.740(3) . ?
S2 C4 1.751(3) . ?
S3 C2 1.739(3) . ?
S3 C5 1.745(3) . ?
S4 C2 1.740(3) . ?

S4 C6 1.749(3) . ?
S5 C3 1.753(3) . ?
S5 C7 1.796(3) . ?
S6 C4 1.745(3) . ?
S6 C8 1.824(3) . ?
S7 C5 1.745(3) . ?
S7 C9 1.800(4) . ?
S8 C6 1.748(3) . ?
S8 C10 1.808(4) . ?
C1 C2 1.363(4) . ?
C3 C4 1.348(4) . ?
C5 C6 1.345(4) . ?
C7 C8 1.514(4) . ?
C7 H7A 0.9700 . ?
C7 H7B 0.9700 . ?
C8 H8A 0.9700 . ?
C8 H8B 0.9700 . ?
C9 C10 1.494(6) . ?
C9 H9A 0.9700 . ?
C9 H9B 0.9700 . ?
C10 H10A 0.9700 . ?
C10 H10B 0.9700 . ?
S11 C11 1.741(3) . ?
S11 C13 1.749(2) . ?
S12 C11 1.738(2) . ?
S12 C14 1.747(3) . ?
S13 C12 1.741(3) . ?
S13 C15 1.744(3) . ?
S14 C12 1.738(3) . ?
S14 C16 1.748(3) . ?
S15 C13 1.740(3) . ?
S15 C17 1.766(5) . ?
S15 C17B 1.878(12) . ?
S16 C18B 1.705(10) . ?
S16 C14 1.739(2) . ?
S16 C18 1.868(5) . ?
S17 C19B 1.714(14) . ?
S17 C15 1.739(3) . ?
S17 C19 1.829(7) . ?
S18 C16 1.751(3) . ?
S18 C20 1.776(4) . ?
S18 C20B 1.862(11) . ?
C11 C12 1.363(3) . ?
C13 C14 1.354(4) . ?
C15 C16 1.355(4) . ?
C17 C18 1.508(7) . ?
C17 H17A 0.9700 . ?
C17 H17B 0.9700 . ?
C18 H18A 0.9700 . ?
C18 H18B 0.9700 . ?
C19 C20 1.508(7) . ?
C19 H19A 0.9700 . ?

C19 H19B 0.9700 . ?
C20 H20A 0.9700 . ?
C20 H20B 0.9700 . ?
C17B C18B 1.521(13) . ?
C17B H17C 0.9700 . ?
C17B H17D 0.9700 . ?
C18B H18C 0.9700 . ?
C18B H18D 0.9700 . ?
C19B C20B 1.508(13) . ?
C19B H19C 0.9700 . ?
C19B H19D 0.9700 . ?
C20B H20C 0.9700 . ?
C20B H20D 0.9700 . ?
Ru1 O1 2.029(2) . ?
Ru1 O1 2.029(2) 2 ?
Ru1 O2 2.050(2) 2 ?
Ru1 O2 2.050(2) . ?
Ru1 O3 2.050(2) 2 ?
Ru1 O3 2.050(2) . ?
K1 O4 2.761(3) 4_455 ?
K1 O4 2.761(3) 3 ?
K1 O6 2.925(3) . ?
K1 O6 2.925(3) 2 ?
K1 O5 2.956(3) 4 ?
K1 O5 2.956(3) 3_455 ?
C21 O4 1.214(4) . ?
C21 O1 1.293(4) . ?
C21 C22 1.555(4) 2 ?
C22 O5 1.227(4) . ?
C22 O2 1.293(4) . ?
C22 C21 1.555(4) 2 ?
C23 O6 1.223(4) . ?
C23 O3 1.280(4) . ?
C23 C23 1.578(7) 2 ?
O4 K1 2.761(3) 3_445 ?
O5 K1 2.956(3) 3_545 ?
Br1 Br1 0.452(7) 2_655 ?
Br1 C24 1.893(7) . ?
Br1 C24 1.921(8) 2_655 ?
C24 C29 1.36(3) . ?
C24 C25 1.41(3) . ?
C25 C26 1.42(2) . ?
C25 H25 0.9300 . ?
C26 C27 1.354(15) . ?
C26 H26 0.9300 . ?
C27 C28 1.401(14) . ?
C27 H27 0.9300 . ?
C28 C29 1.36(3) . ?
C28 H28 0.9300 . ?
C29 H29 0.9300 . ?

loop_

```
 _geom_angle_atom_site_label_1
 _geom_angle_atom_site_label_2
 _geom_angle_atom_site_label_3
 _geom_angle
 _geom_angle_site_symmetry_1
 _geom_angle_site_symmetry_3
 _geom_angle_publ_flag
C1 S1 C3 95.72(12) . . ?
C1 S2 C4 95.29(12) . . ?
C2 S3 C5 95.47(12) . . ?
C2 S4 C6 95.46(13) . . ?
C3 S5 C7 101.69(13) . . ?
C4 S6 C8 101.63(13) . . ?
C5 S7 C9 99.23(16) . . ?
C6 S8 C10 101.95(17) . . ?
C2 C1 S2 123.0(2) . . ?
C2 C1 S1 121.99(19) . . ?
S2 C1 S1 115.05(15) . . ?
C1 C2 S3 122.1(2) . . ?
C1 C2 S4 123.0(2) . . ?
S3 C2 S4 114.96(14) . . ?
C4 C3 S1 116.6(2) . . ?
C4 C3 S5 128.5(2) . . ?
S1 C3 S5 114.95(14) . . ?
C3 C4 S6 128.7(2) . . ?
C3 C4 S2 117.4(2) . . ?
S6 C4 S2 113.96(15) . . ?
C6 C5 S7 127.8(2) . . ?
C6 C5 S3 117.2(2) . . ?
S7 C5 S3 115.00(16) . . ?
C5 C6 S8 129.0(2) . . ?
C5 C6 S4 116.9(2) . . ?
S8 C6 S4 114.02(16) . . ?
C8 C7 S5 114.5(2) . . ?
C8 C7 H7A 108.6 . . ?
S5 C7 H7A 108.6 . . ?
C8 C7 H7B 108.6 . . ?
S5 C7 H7B 108.6 . . ?
H7A C7 H7B 107.6 . . ?
C7 C8 S6 114.0(2) . . ?
C7 C8 H8A 108.8 . . ?
S6 C8 H8A 108.8 . . ?
C7 C8 H8B 108.8 . . ?
S6 C8 H8B 108.8 . . ?
H8A C8 H8B 107.6 . . ?
C10 C9 S7 113.1(3) . . ?
C10 C9 H9A 109.0 . . ?
S7 C9 H9A 109.0 . . ?
C10 C9 H9B 109.0 . . ?
S7 C9 H9B 109.0 . . ?
H9A C9 H9B 107.8 . . ?
C9 C10 S8 114.9(3) . . ?
```

C9 C10 H10A 108.5 . . ?
S8 C10 H10A 108.5 . . ?
C9 C10 H10B 108.5 . . ?
S8 C10 H10B 108.5 . . ?
H10A C10 H10B 107.5 . . ?
C11 S11 C13 95.48(12) . . ?
C11 S12 C14 94.86(12) . . ?
C12 S13 C15 95.05(12) . . ?
C12 S14 C16 95.73(12) . . ?
C13 S15 C17 106.21(19) . . ?
C13 S15 C17B 94.1(4) . . ?
C18B S16 C14 108.3(3) . . ?
C14 S16 C18 95.87(18) . . ?
C19B S17 C15 105.0(5) . . ?
C15 S17 C19 100.1(3) . . ?
C16 S18 C20 102.93(16) . . ?
C16 S18 C20B 96.5(3) . . ?
C12 C11 S12 122.6(2) . . ?
C12 C11 S11 122.0(2) . . ?
S12 C11 S11 115.31(14) . . ?
C11 C12 S14 122.9(2) . . ?
C11 C12 S13 121.8(2) . . ?
S14 C12 S13 115.23(14) . . ?
C14 C13 S15 127.46(19) . . ?
C14 C13 S11 116.20(19) . . ?
S15 C13 S11 116.28(15) . . ?
C13 C14 S16 125.7(2) . . ?
C13 C14 S12 117.76(19) . . ?
S16 C14 S12 116.39(15) . . ?
C16 C15 S17 128.1(2) . . ?
C16 C15 S13 117.80(19) . . ?
S17 C15 S13 114.09(14) . . ?
C15 C16 S14 116.15(19) . . ?
C15 C16 S18 128.0(2) . . ?
S14 C16 S18 115.79(14) . . ?
C18 C17 S15 116.3(4) . . ?
C18 C17 H17A 108.2 . . ?
S15 C17 H17A 108.2 . . ?
C18 C17 H17B 108.2 . . ?
S15 C17 H17B 108.2 . . ?
H17A C17 H17B 107.4 . . ?
C17 C18 S16 113.2(4) . . ?
C17 C18 H18A 108.9 . . ?
S16 C18 H18A 108.9 . . ?
C17 C18 H18B 108.9 . . ?
S16 C18 H18B 108.9 . . ?
H18A C18 H18B 107.7 . . ?
C20 C19 S17 113.3(5) . . ?
C20 C19 H19A 108.9 . . ?
S17 C19 H19A 108.9 . . ?
C20 C19 H19B 108.9 . . ?
S17 C19 H19B 108.9 . . ?

H19A C19 H19B 107.7 . . ?
C19 C20 S18 113.1(4) . . ?
C19 C20 H20A 109.0 . . ?
S18 C20 H20A 109.0 . . ?
C19 C20 H20B 109.0 . . ?
S18 C20 H20B 109.0 . . ?
H20A C20 H20B 107.8 . . ?
C18B C17B S15 113.4(7) . . ?
C18B C17B H17C 108.9 . . ?
S15 C17B H17C 108.9 . . ?
C18B C17B H17D 108.9 . . ?
S15 C17B H17D 108.9 . . ?
H17C C17B H17D 107.7 . . ?
C17B C18B S16 114.1(8) . . ?
C17B C18B H18C 108.7 . . ?
S16 C18B H18C 108.7 . . ?
C17B C18B H18D 108.7 . . ?
S16 C18B H18D 108.7 . . ?
H18C C18B H18D 107.6 . . ?
C20B C19B S17 113.4(8) . . ?
C20B C19B H19C 108.9 . . ?
S17 C19B H19C 108.9 . . ?
C20B C19B H19D 108.9 . . ?
S17 C19B H19D 108.9 . . ?
H19C C19B H19D 107.7 . . ?
C19B C20B S18 113.8(9) . . ?
C19B C20B H20C 108.8 . . ?
S18 C20B H20C 108.8 . . ?
C19B C20B H20D 108.8 . . ?
S18 C20B H20D 108.8 . . ?
H20C C20B H20D 107.7 . . ?
O1 Ru1 O1 90.74(14) . 2 ?
O1 Ru1 O2 81.36(9) . 2 ?
O1 Ru1 O2 90.81(9) 2 2 ?
O1 Ru1 O2 90.81(9) . . ?
O1 Ru1 O2 81.36(9) 2 . ?
O2 Ru1 O2 168.89(14) 2 . ?
O1 Ru1 O3 174.36(10) . 2 ?
O1 Ru1 O3 94.40(10) 2 2 ?
O2 Ru1 O3 96.21(10) 2 2 ?
O2 Ru1 O3 92.26(10) . 2 ?
O1 Ru1 O3 94.40(10) . . ?
O1 Ru1 O3 174.36(10) 2 . ?
O2 Ru1 O3 92.26(10) 2 . ?
O2 Ru1 O3 96.21(10) . . ?
O3 Ru1 O3 80.57(14) 2 . ?
O4 K1 O4 136.97(13) 4_455 3 ?
O4 K1 O6 103.76(8) 4_455 . ?
O4 K1 O6 114.06(8) 3 . ?
O4 K1 O6 114.06(8) 4_455 2 ?
O4 K1 O6 103.76(8) 3 2 ?
O6 K1 O6 56.66(11) . 2 ?

O4 K1 O5 92.52(8) 4_455 4 ?
O4 K1 O5 57.47(7) 3 4 ?
O6 K1 O5 158.94(9) . 4 ?
O6 K1 O5 104.60(8) 2 4 ?
O4 K1 O5 57.47(7) 4_455 3_455 ?
O4 K1 O5 92.52(8) 3 3_455 ?
O6 K1 O5 104.60(8) . 3_455 ?
O6 K1 O5 158.94(9) 2 3_455 ?
O5 K1 O5 95.36(12) 4 3_455 ?
O4 C21 O1 125.5(3) . . ?
O4 C21 C22 120.2(3) . 2 ?
O1 C21 C22 114.2(3) . 2 ?
O5 C22 O2 125.9(3) . . ?
O5 C22 C21 118.2(3) . 2 ?
O2 C22 C21 115.9(3) . 2 ?
O6 C23 O3 125.9(4) . . ?
O6 C23 C23 119.3(2) . 2 ?
O3 C23 C23 114.8(2) . 2 ?
C21 O1 Ru1 114.77(19) . . ?
C22 O2 Ru1 112.84(18) . . ?
C23 O3 Ru1 114.9(2) . . ?
C21 O4 K1 124.9(2) . 3_445 ?
C22 O5 K1 118.5(2) . 3_545 ?
C23 O6 K1 122.3(3) . . ?
Br1 Br1 C24 86.8(7) 2_655 . ?
Br1 Br1 C24 79.6(7) 2_655 2_655 ?
C29 C24 C25 123.5(8) . . ?
C29 C24 Br1 116.5(16) . . ?
C25 C24 Br1 119.9(16) . . ?
C24 C25 C26 116.1(13) . . ?
C24 C25 H25 121.9 . . ?
C26 C25 H25 122.0 . . ?
C27 C26 C25 119.9(12) . . ?
C27 C26 H26 120.0 . . ?
C25 C26 H26 120.0 . . ?
C26 C27 C28 121.7(8) . . ?
C26 C27 H27 119.2 . . ?
C28 C27 H27 119.2 . . ?
C29 C28 C27 120.1(13) . . ?
C29 C28 H28 119.9 . . ?
C27 C28 H28 120.0 . . ?
C28 C29 C24 118.6(14) . . ?
C28 C29 H29 120.7 . . ?
C24 C29 H29 120.7 . . ?

Table S1. $t_{HOMO-HOMO}$ transfer integrals (meV) and S···S distances shorter than 3.9 Å for the different donor···donor interactions in β''-(BEDT-TTF)$_4$K$_x$(H$_3$O)$_{1-x}$[Ru^{III}(ox)$_3$]C$_6$H$_5$Br (x = 0.77, sample #4 with minimal T_c).

Interaction		S···S (<3.9Å)	$t_{HOMO-HOMO}$ (meV)
I	(A-B)	3.661, 3.796, 3.828, 3.865	-142
II	(A-A)	3.738(x2)	-50
III	(B-B)	3.823	-57
IV	(A-B)	3.602, 3.734, 3.739, 3.798	+205
V	(A-B)	3.594, 3.766, 3.773, 3.887	+180
VI	(B-B)	3.408(x2),3.523(x2), 3.846, 3.847(x2)	-97
VII	(A-B)	3.322, 3.380, 3.530, 3.563,3.841	-95
VIII	(A-A)	3.666(x2), 3.859	-55

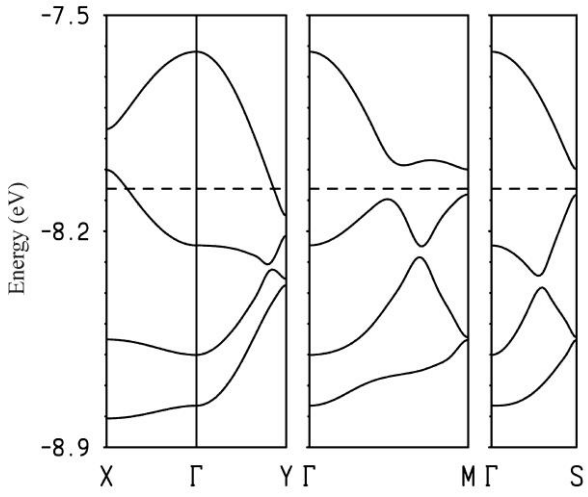

Figure S3. Band structure calculated for a donor layer of β''-(BEDT-TTF)$_4$K$_x$(H$_3$O)$_{1-x}$[Ru^{III}(ox)$_3$]C$_6$H$_5$Br (sample #4 with minimal T_c). The dashed line refers to the Fermi level. Γ = (0, 0), X = (*a*'*/2, 0), Y = (0. *b*'*/2). M = (*a*'*/2, *b*'*/2) and S = (-*a*'*/2, *b*'*/2) where a'= (*a*/2 - *b*/2) and *b*' = (*a*/2 + *b*/2).

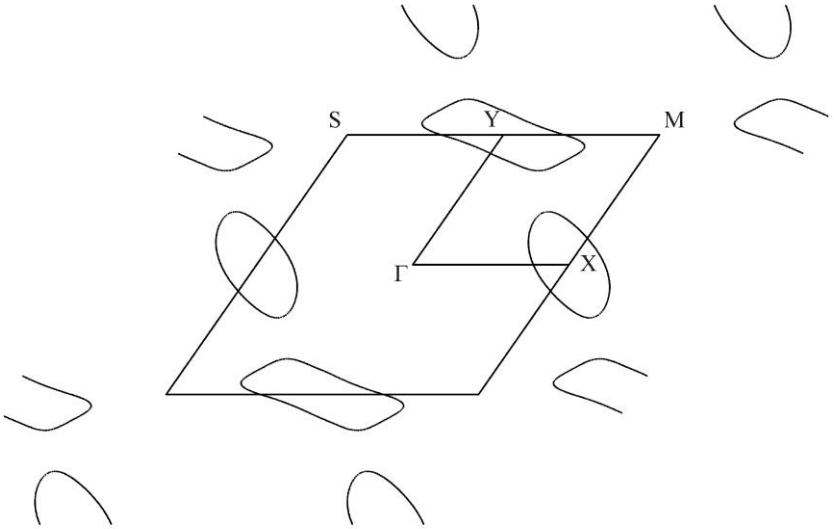

Figure S4. Fermi surface calculated for a donor layer of β''-(BEDT-TTF)$_4$K$_x$(H$_3$O)$_{1-x}$[Ru^{III}(ox)$_3$]C$_6$H$_5$Br (sample #4 with minimal T_c).