# KFM 8: Multiferroics and magnetoelectrics I (joint session MA/KFM)

Time: Tuesday 9:30-12:15

KFM 8.1 Tue 9:30 EB 202

Lu2Fe3O7 a quest for ferroelectricity by charge order — •SABREEN HAMMOUDA and MANUEL ANGST — Jülich Centre for Neutron Science JCNS and Peter Grünberg Institut PGI, JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.

Rare earth ferrites have attracted a lot of attention as proposed multiferroics. In particular, LuFe2O4 was considered a clear example of ferroelectricity from charge ordering (CO), though recently this was found not to be the case [1]. Structural modification, such as intercalation by LuFeO3, yielding Lu2Fe3O7 might render the bilayers polar. Furthermore, the only believable polarization hysteresis loop in the rare earth ferrite literature [2] was measured on an intercalated compound, slightly Mn-doped Lu2Fe3O7. A critical aspect of investigating these compounds is the oxygen-stoichiometry. We succeeded in growing single crystals of Lu2Fe3O7 using different CO2/CO gas mixtures to fine-tune the oxygen partial pressure. Single crystals examined by x-ray diffraction showed a short range ordering with a zigzagged diffuse scattering along  $(1/3 \ 1/3 \ 1)$ , with positions similar to the observation by electron diffraction [3]. The diffuse nature indicates that the crystals are not quite stoichiometric enough for long range CO. Powder XRD measurements reveal a peak splitting which is likely due to structural distortion because of CO. Magnetic behavior of these crystals will also be discussed. As an outlook, further optimization is needed to determine the charge and spin structures. [1] de Groot et al., Phys. Rev. Lett. 108, 187601 (2012). [2] Qin et al., Appl. Phys. Lett. 95, 072901 (2009). [3] Yang et al., Phys. Status Solidi B 247, 870 (2010).

### KFM 8.2 Tue 9:45 EB 202

Structural and spectroscopic properties of the new multiferroic  $Ni_2MnTeO_6$  — •Stella Skiadopoulou<sup>1,2</sup>, Maria Retuerto<sup>3</sup>, Fedir Borodavka<sup>1</sup>, Christelle Kadlec<sup>1</sup>, Filip Kadlec<sup>1</sup>, Zheng Deng<sup>3</sup>, Martha Greenblatt<sup>3</sup>, Dominik Legut<sup>2</sup>, and Stanislav Kamba<sup>1</sup> — <sup>1</sup>Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic — <sup>2</sup>VSB Techincal University of Ostrava, Ostrava, Czech Republic — <sup>3</sup>Rutgers, The State University of New Jersey, Piscataway, USA

We present structural, magnetic and spectroscopic studies of a new multiferroic Ni<sub>2</sub>MnTeO<sub>6</sub>, closely related to the polar antiferromagnet Ni<sub>3</sub>TeO<sub>6</sub> known to present a colossal magnetoelectric effect and electromagnons. Single crystals and polycrystalline samples show the same polar structure as Ni<sub>3</sub>TeO<sub>6</sub> with the R3 space group down to 4 K. An antiferromagnetic phase transition takes place at approximately  $T_N=70$  K, almost 20 K higher than that of Ni<sub>3</sub>TeO<sub>6</sub>. This was confirmed by magnetic and dielectric measurements, suggesting the multiferroic character of the compound. Extensive infrared, Raman and THz spectroscopy experiments revealed all phonons predicted by the factor group analysis. THz spectra reveal one new excitation below  $T_N$ , which is strongly influenced by external magnetic field, thus assigned to a magnon.

This work was supported by Czech Science Foundation grant No. 17-27790S and Path to Exascale project No. CZ.02.1.01/0.0/0.0/16\_013/0001791.

KFM 8.3 Tue 10:00 EB 202 Exotic magnetoelectric excitations of the multiferroic SmFe3(BO3)4 — •DÁVID SZALLER<sup>1</sup>, ARTEM M. KUZ'MENKO<sup>2</sup>, ALEXANDER A. MUKHIN<sup>2</sup>, TOOMAS RÕÕM<sup>3</sup>, URMAS NAGEL<sup>3</sup>, THOMAS KAIN<sup>1</sup>, VLAD DZIOM<sup>1</sup>, LUKAS WEYMANN<sup>1</sup>, ALEXEY SHUVAEV<sup>1</sup>, ANNA PIMENOV<sup>1</sup>, VSEVOLOD YU. IVANOV<sup>2</sup>, IRINA A. GUDIM<sup>4</sup>, LEONARD N. BEZMATERNYKH<sup>4</sup>, and ANDREI PIMENOV<sup>1</sup> — <sup>1</sup>Institute of Solid State Physics, Vienna University of Technology, 1040 Vienna, Austria — <sup>2</sup>Prokhorov General Physics Institute, Russian Academy of Sciences, 119991 Moscow, Russia — <sup>3</sup>National Institute of Chemical Physics and Biophysics, Akadeemia tee 23, 12618 Tallinn, Estonia — <sup>4</sup>L. V. Kirensky Institute of Physics Siberian Branch of RAS, 660036 Krasnoyarsk, Russia

Magnetoelectric(ME) multiferroics(MFs), i.e. materials simultaneously hosting ferroelectric and magnetic order, have been attracting enormous interest due to their potential in information-technology applications. Rare-earth ferroborates are a particularly interesting familiy of MF crystals, where the strong spin-orbit interaction at the rare-earth sites results in the coupling of the magnetic and electric degrees of freedom while the ME response is enhanced by the antiferromagnetic ordering of the iron spins. The ME coupling appears in the optical regime as different absorption of counter-propagating light beams, where transparent and dark directions can be swapped by reversing the magnetic field. Furthermore, due to the ME coupling the strength of absorption at spin-wave resonance frequencies can also be tuned by electric field, opening the path for practical applications.

Recently, it was found that the polar ferrimagnet  $Mn_2Mo_3O_8$  shows large diagonal magnetoelectric effect [1]. However, the magnetic structure of this compound is not fully understood. The family of polar ferrimagnets  $M_2Mo_3O_8$  with M = Mn, Fe, Co or Ni are excellent materials to investigate the role of different magnetic ions in the microscopic origin of magnetoelectric effect.

In this work magnetization measurements and high magnetic field far infrared spectroscopy are used to unreveal the spin excitations in the low-field ferrimagnetic and in the spin-flop phases. From the magnetic field dependence of the magnon excitation energies we aim to determine the most important exchange and anisotropy parameters. With these parameters it is possible to construct a microscopic spin model of this compound.

[1] T. Kurumaji et al.: PRB 95, 045142 (2017).

KFM 8.5 Tue 10:30 EB 202 Local Magnetic and Electric Interactions in Multiferroic Ba<sub>2</sub>CoGe<sub>2</sub>O<sub>7</sub> and Sr<sub>2</sub>CoSi<sub>2</sub>O<sub>7</sub> — •MARTINA SCHÄDLER<sup>1</sup>, TI-TUSZ FEHÉR<sup>2</sup>, NORBERT BÜTTGEN<sup>1</sup>, VILMOS KOCSIS<sup>3</sup>, YOSHINORI TOKURA<sup>3</sup>, YASUJIRO TAGUCHI<sup>3</sup>, and ISTVÁN KÉZSMÁRKI<sup>1</sup> — <sup>1</sup>Center for Electronic Correlations and Magnetism, Institute of Physics, University of Augsburg, Germany — <sup>2</sup>Department of Physics, Budapest University of Technology and Economics, Hungary — <sup>3</sup>RIKEN Center for Emergent Matter Science, Wako 351-0198, Japan

The multiferroic compound Ba<sub>2</sub>CoGe<sub>2</sub>O<sub>7</sub> has drawn a lot of interest due to its non-centrosymmetric crystal structure, giving rise to peculiar magnetoelectric effects. The spin-dependent hybridization mechanism, that induces the electric polarization, results in a strong coupling of the magnetic moments and the local electric polarization, which allows control of the electric polarization via external magnetic fields. Due to its soft antiferromagnetic structure Ba<sub>2</sub>CoGe<sub>2</sub>O<sub>7</sub> is a promising candidate for tuning the magnetic texture via the application of electric fields. Nuclear Magnetic Resonance (NMR) gives access to the local electric field gradient (EFG) via the nuclear quadrupole moment. We performed <sup>59</sup>Co NMR measurements on Ba<sub>2</sub>CoGe<sub>2</sub>O<sub>7</sub> and its sister compound Sr<sub>2</sub>CoSi<sub>2</sub>O<sub>7</sub> in order to determine the local microscopic properties of magnetic spin order and electric polarization at the cobalt site. Through additional application of external electric fields we also investigated the possibility of influencing the local magnetic properties.

#### 15 minutes break

KFM 8.6 Tue 11:00 EB 202 Soft modes in Ca<sub>3</sub>Mn<sub>2</sub>O<sub>7</sub> - Direct observation of the order parameters in a hybrid improper ferroelectric material — •DIRK WULFERDING<sup>1,2</sup>, ALEXANDER GLAMAZDA<sup>3,1</sup>, PETER LEMMENS<sup>1,2</sup>, BIN GAO<sup>4</sup>, SANG-WOOK CHEONG<sup>4</sup>, and KWANG-YONG CHOI<sup>5</sup> — <sup>1</sup>IPKM, TU-BS, Braunschweig, Germany — <sup>2</sup>LENA, TU-BS, Braunschweig, Germany — <sup>3</sup>ILTPE, NASU, Kharkov, Ukraine — <sup>4</sup>Rutgers Univ., New Jersey, USA — <sup>5</sup>Chung-Ang Univ., Seoul, Korea In hybrid improper ferroelectric materials the order parameter is still under debate, but predicted to be a combination of rotation and tilting modes. In the title compound Ca<sub>3</sub>Mn<sub>2</sub>O<sub>7</sub> we observe anomalous softening of rotation and tilting phonons through the transition from

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the ferroelectric to the paraelectric phase. This clearly underlines their role as order parameters. In addition, a coupling of the soft mode to the magnetic and the electronic subsystems is characterized through an observation of anomalous magnetic and multiphonon Raman scattering. Work supported by the Quantum- and Nanometrology initiative "QUANOMET" within project NL-4, DFG-RTG 1952/1 "NanoMet", Korea NRF Grants (No. 2009-0093817, 2012-046138), and the NSF MRI Grant No. MRI-1532006.

#### KFM 8.7 Tue 11:15 EB 202

Giant magnetoelectric coupling in the low-dimensional ferrimagnetic iron oxoselenite  $Fe_2O(SeO_3)_2 - \bullet PETER$ LEMMENS<sup>1,2</sup>, VLADIMIR GNEZDILOV<sup>3</sup>, DIRK WULFERDING<sup>1,2</sup>, PETER BERDONOSOV<sup>4</sup>, E.S. KOZLYAKOVA<sup>4</sup>, E. KUZNETSOVA<sup>4</sup>, OLGA VOLKOVA<sup>4</sup>, and ALEXANDER VASILIEV<sup>4</sup> - <sup>1</sup>IPKM, TU-BS, Braunschweig, Germany - <sup>2</sup>LENA, TU-BS, Braunschweig, Germany - <sup>3</sup>ILTPE, NASU, Kharkov, Ukraine - <sup>4</sup>MSU, Moscow, Russia

The newly synthesized oxoselenite compound Fe<sub>2</sub>O(SeO<sub>3</sub>)<sub>2</sub> hosts Fe ions in distorted tetrahedral coordination. An anomalous gain in phonon intensity observed within the ferromagnetically ordered phase ( $T_C = 105$  K) hints towards an enhanced electronic polarizability and related giant magnetoelectric coupling. Further anomalous modes of possible magnetic origin are discussed in connection with a strong Dzyaloshinskii-Moriya interaction. Work supported by DFG Project LE967/16-1.

KFM 8.8 Tue 11:30 EB 202

Magnetic and Polar Properties of the Lacunar Spinel  $GaMo_4S_8 - \bullet$ KORBINIAN GEIRHOS<sup>1</sup>, PETER LUNKENHEIMER<sup>1</sup>, HI-ROYUKI NAKAMURA<sup>2</sup>, YOSHIKAZU TABATA<sup>2</sup>, and ISTVÁN KÉZSMÁRKI<sup>1</sup> -<sup>1</sup>Experimental Physics V, EKM, University of Augsburg, Germany

- <sup>2</sup>Department of Materials Science and Engineering, Kyoto University, Japan

The compound GaMo<sub>4</sub>S<sub>8</sub> belongs to the family of lacunar spinels  $AM_4X_8$  (A=Ga and Ge; M=V, Mo, Nb, and Ta; X= S and Se). Many of these lacunar spinels exhibit a Jahn-Teller transition associated with ferroorbital ordering. In the so far investigated compounds  $GaV_4S_8$ ,  $GaV_4Se_8$  and  $GeV_4S_8$ , the onset of orbital-order induced ferroelectricity was found at the Jahn-Teller transition [1,2,3]. Moreover, all of these three materials show strong magnetoelectric coupling with distinct values of the polarization in their magnetically ordered phases, including a skyrmion lattice state, as shown for  $GaV_4S_8$  and  $GaV_4Se_8$  [1, 4]. It was proposed that these skyrmions, which are topologically protected spin textures, carry additional electric polarization [1]. We extend these investigations to another lacunar spinel, GaMo<sub>4</sub>S<sub>8</sub>. The polar properties of this compound were studied by dielectric spectroscopy and pyrocurrent measurements. It shows a Jahn-Teller transition at 47 K, again accompanied by polar ordering. In addition, below 20 K GaMo<sub>4</sub>S<sub>8</sub> exhibits a complex magnetic phase diagram.

[1] E. Ruff et al., Sci. Adv. 1, E1500916 (2015). [2] E. Ruff et al., Phys. Rev. B 96, 165119 (2017). [3] K. Singh et al., Phys. Rev. Lett.
113, 137602 (2014). [4] Y. Fujima et al., Phys. Rev. B 95, 180410 (2017)

## KFM 8.9 Tue 11:45 EB 202

Ferrimagnetic-type in-gap spin excitations and magnetoelastic coupling in  $\alpha$ -Cu<sub>2</sub>V<sub>2</sub>O<sub>7</sub> — •JOHANNES WERNER<sup>1</sup>, LIRAN WANG<sup>1</sup>, ALEXANDER OTTMANN<sup>1</sup>, ROBIN WEIS<sup>1</sup>, MAHMOUD ABDEL-HAFIEZ<sup>2</sup>, JHUMA SANNIGRAHI<sup>3</sup>, SOURADIP MAJUMDAR<sup>4</sup>, CHANGHYUN KOO<sup>1</sup>, and RÜDIGER KLINGELER<sup>1</sup> — <sup>1</sup>Kirchhoff Institute for Physics, Heidelberg, Germany — <sup>2</sup>Physikalisches Institut, Goethe-Universität, Frankfurt a.M., Germany — <sup>3</sup>ISIS Facility, Rutherford Appleton Laboratory, Didcot, United Kingdom — <sup>4</sup>Department of Solid State Physics, Kolkata, India

Low-energy magnetic excitations and magnetoelastic coupling in multiferroic  $\alpha$ -Cu<sub>2</sub>V<sub>2</sub>O<sub>7</sub> have been investigated by high-frequency electron spin resonance (HF-ESR) , thermal expansion, magnetostriction, specific heat and magnetisation studies in magnetic fields up to 15 T. Despite a large antiferromagnetic gap, below 100 GHz we observe low-energy magnetic excitations in the spin ordered phase indicating a ferrimagnetic-type resonance branch associated with the Dzyaloshinsii-Moriya-type canted magnetic moment. The anisotropy parameter  $\tilde{D} = 1.3(1)$  meV indicates a sizeable ratio of DM-exchange and magnetic anisotropy. Dilatometry results show negative thermal expansion at T  $\leq$  200 K. Pronounced anomalies at  $T_{\rm N}$  = 35 K imply coupling to the structure. Failure of Grüneisen scaling confirm that several ordering phenomena are concomitantly driving the multiferroic order. Low-field magnetostriction displays a similar hysteresis loop as the magnetisation which supports the scenario of exchange-striction driven multiferroicity.

KFM 8.10 Tue 12:00 EB 202 Directional dichroism via para-magnetoelectric effect in  $Sr_2CoSi_2O_7 - \bullet$ Dániel G. Farkas<sup>1</sup>, Dávid Szaller<sup>1</sup>, Vilmos Kocsis<sup>1,2</sup>, Sándor Bordács<sup>1</sup>, István Kézsmárki<sup>1</sup>, Bence Bernáth<sup>3</sup>, Dmytro Kamenskyi<sup>3</sup>, Laur Peedu<sup>4</sup>, Johan Viirok<sup>4</sup>, Toomas Rõõm<sup>4</sup>, Urmas Nagel<sup>4</sup>, Péter Balla<sup>5</sup>, and Karlo Penc<sup>5</sup> - <sup>1</sup>BUTE, Hungary - <sup>2</sup>RIKEN CEMS, Japan - <sup>3</sup>HFML, Netherlands - <sup>4</sup>KBFI, Estonia - <sup>5</sup>WRCP, Hungary

Magnetoelectric multiferroics have been attracting enormous interest due to their potential in information technology applications. An exotic phenomenon, directional dichroism (DD) has been reported for spin excitations in multiferroic melilite single crystals and proposed as a new principle of directional light switches operating in the THz region [1].

Applications of multiferroic compounds seem to be limited to low temperatures where electric and magnetic order coexist. However, recent studies on melilites [2] revealed that an external magnetic field can recover the electric polarization via the para-magnetoelectric effect even above  $T_N = 7$  K. Based on these static results we can also expect DD to emerge in the paramagnetic phase of melilites.

Indeed we have found strong DD in the paramagnetic phase of  $Sr_2CoSi_2O_7$  in high magnetic field. A simple single-ion model was developed, which described the main features of the high temperature directional dichroism.

I. Kézsmárki, D. Szaller et al. Nat. Commun. 5, 3203 (2013).
 M. Akaki et al., Phys. Rev. B 86, 060413(R) (2012).