IWAMO
2nd International Workshop on Advanced Magnetic Oxides
2021

Engineering of Lanthanum Perovskite Thin Films for Magnetic Sensors Applications

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Outline

- Introduction: increasing market of magnetoresistive sensors
- Lanthanum manganites: nanostructured films exhibiting negative colossal magnetoresistance (CMR)
- Engineering of manganite films: influence of substrate, chemical composition, film thickness and deposition temperature
- CMR-B-scalar magnetic sensors: measurement of high pulsed magnetic fields
- 2D materials: Graphene exhibiting large positive magnetoresistance
- Novel prototype: Hybrid magnetoresistive manganite/graphene sensor
- Summary and future perspectives

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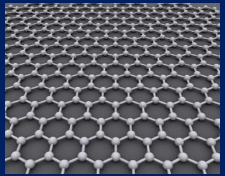

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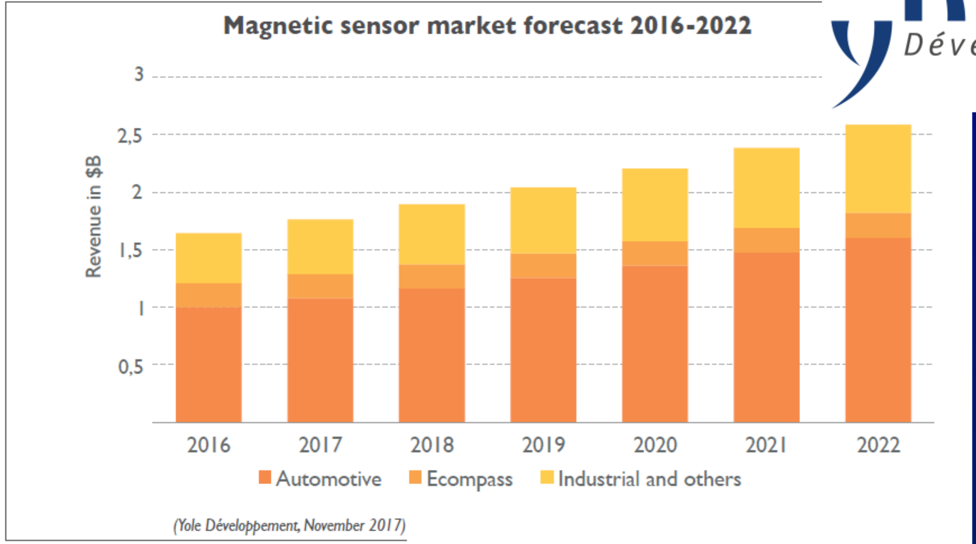
Growing market of magnetic sensors based on xMR (AMR, GMR, TMR) technologies

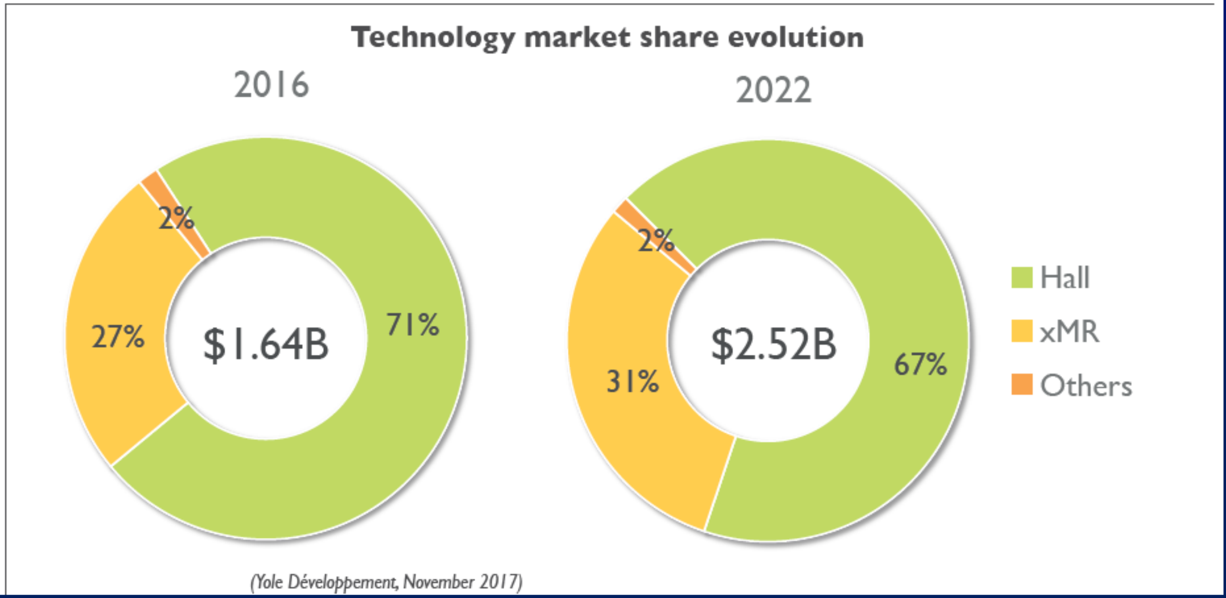
http://www.yole.fr/Magnetic_Sensor_Market.aspx#.WmoQO3mLlaQ



THE COMPETITIVE LANDSCAPE VARIES DEPENDING ON WHETHER THE MARKET IS FOCUSED ON AUTOMOTIVE, ECOMPASS OR INDUSTRIAL AND OTHERS

xMR TECHNOLOGIES WILL SLOWLY GAIN MARKET SHARE

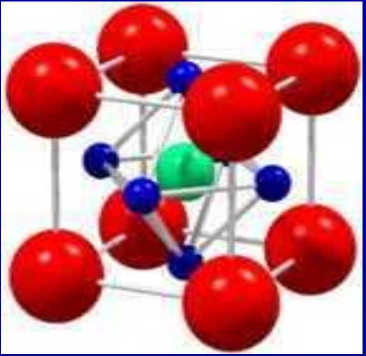
The technology landscape is becoming increasingly complex, as the presence of magnetoresistive (xMR) technologies grows. These include anisotropic magnetoresistive (AMR), giant magnetoresistive (GMR), and more recently tunnel magnetoresistive (TMR). Their growth comes at the expense of, or in combination with, historical Hall effect technology. xMR technologies' main advantage is better sensitivity and thus they are mainly used for ecompass and position measurement, especially for precise angles. Most big Hall players have introduced xMR technologies into their product portfolios and intend to grow this business, including AKM, Infineon, Allegro, Melexis, TDK, Diodes and Honeywell. Thus, we expect xMR technologies will increase their market share from 27% to 31% from 2016 to 2022.



The increasing demand in magnetic field sensors with increased sensitivity and decreased dimensions has resulted in extensive investigations of magnetoresistive effects called as *xMR* and search of new materials and nanostructures.

Colossal MagnetoResistive (CMR) effect ?

https://www.sensitec.com/fileadmin/sensitec/Service_and_Support/Downloads/Catalogue/Sensitec_Katalog2015_Web_LZen.pdf



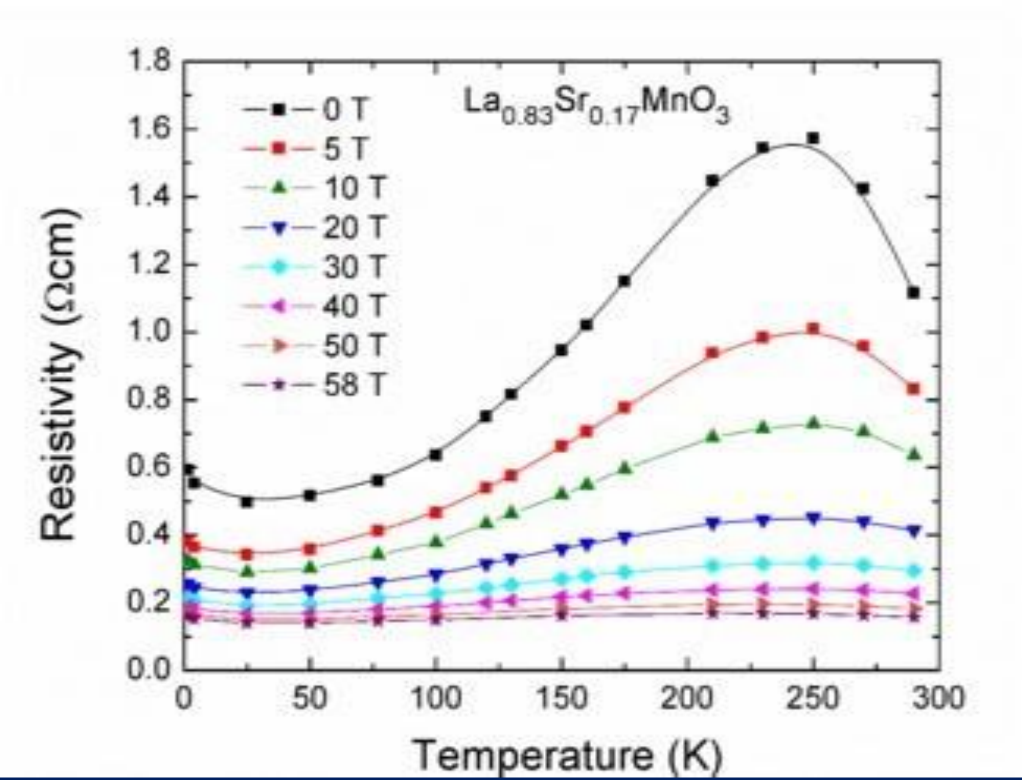
Colossal Magnetoresistance (CMR) materials: lanthanum manganite films

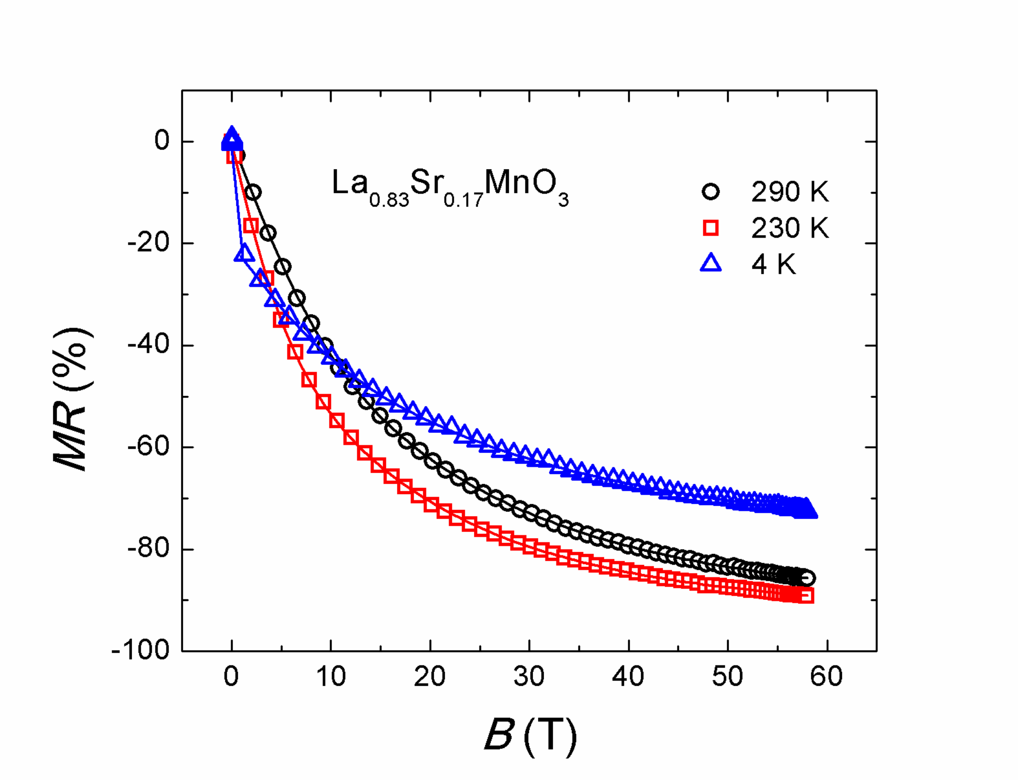
A-site doping — **B-site doping**

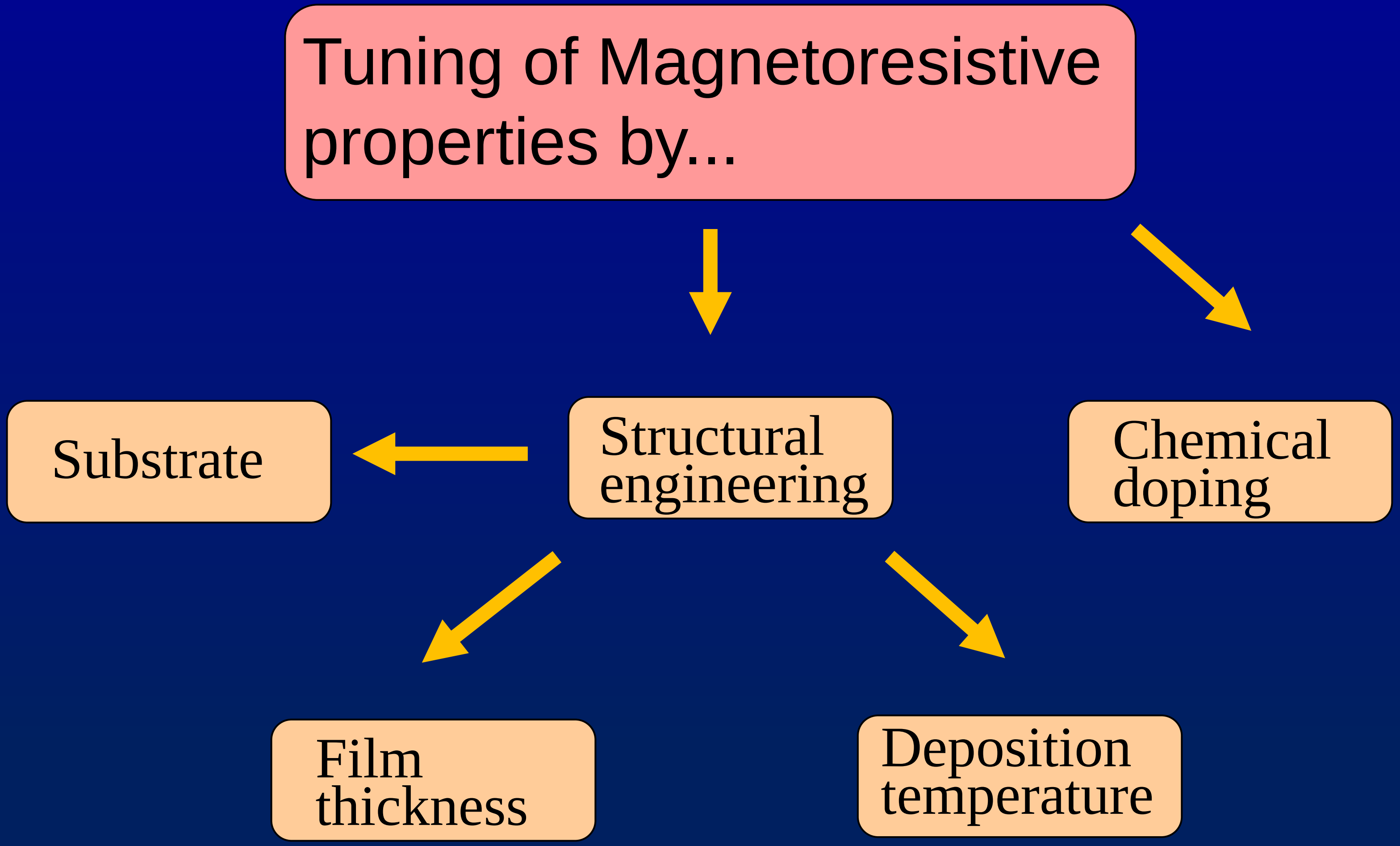
ABO_3 — **$La_{1-x}Sr_xMnO_3$** — **$La_{1-x}Sr_x(Mn_{1-y}Co_y)_zO_3$**

Colossal magnetoresistance (negative)

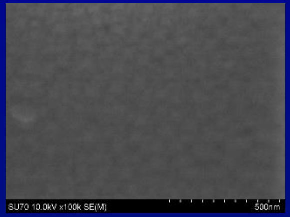
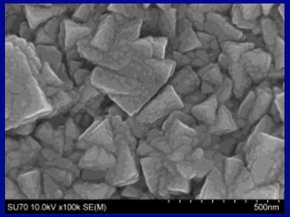
$$MR = 100 \times \frac{\rho(B) - \rho(0)}{\rho(0)}$$

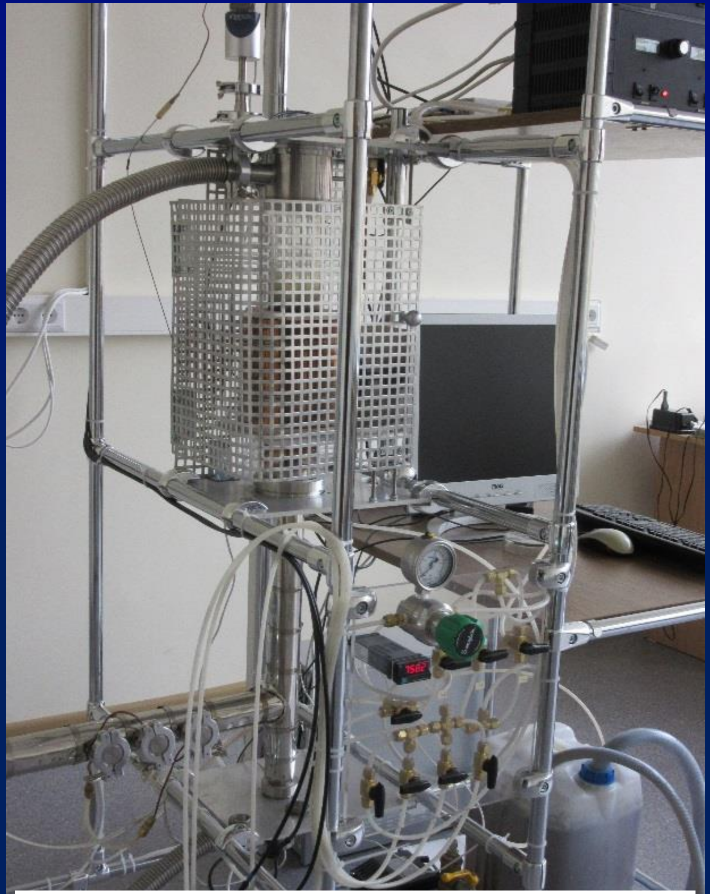





Tuning of Magnetoresistive properties by...
Substrate
Structural engineering
Chemical doping
Film thickness
Deposition temperature

Pulsed Injection Metal–Organic Chemical Vapor Deposition (PI MOCVD)



Vilnius University production

Deposition of epitaxial and polycrystalline thin films of **ferromagnetic oxides** on various substrates

Deposition temperatures:
600 °C - 825 °C

Film thickness:
4 - 500 nm epitaxial films;
20-900 nm polycrystalline films.

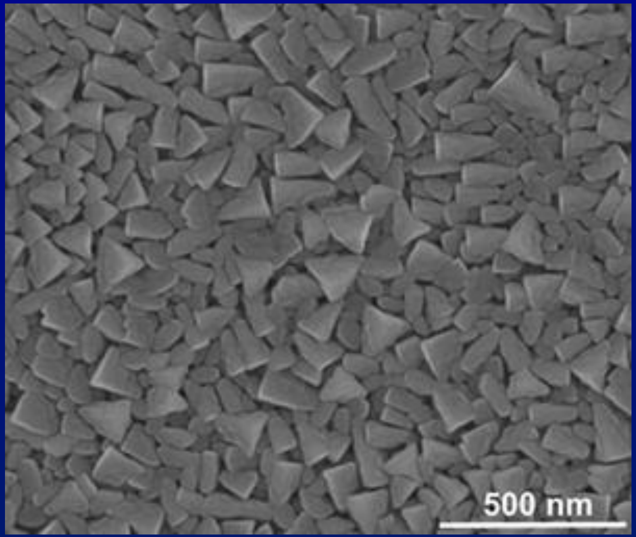
Substrates: $LaAlO_3$, $SrTiO_3$, $NdGaO_3$, Al_2O_3, glass-ceramics, etc.

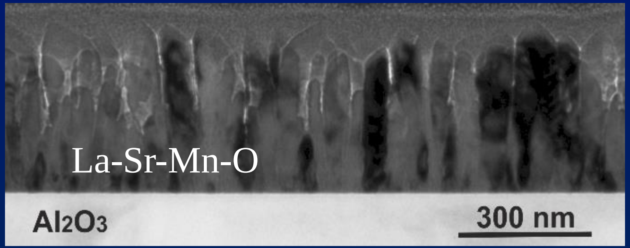
$La_{1-x}Sr_xMnO_3$

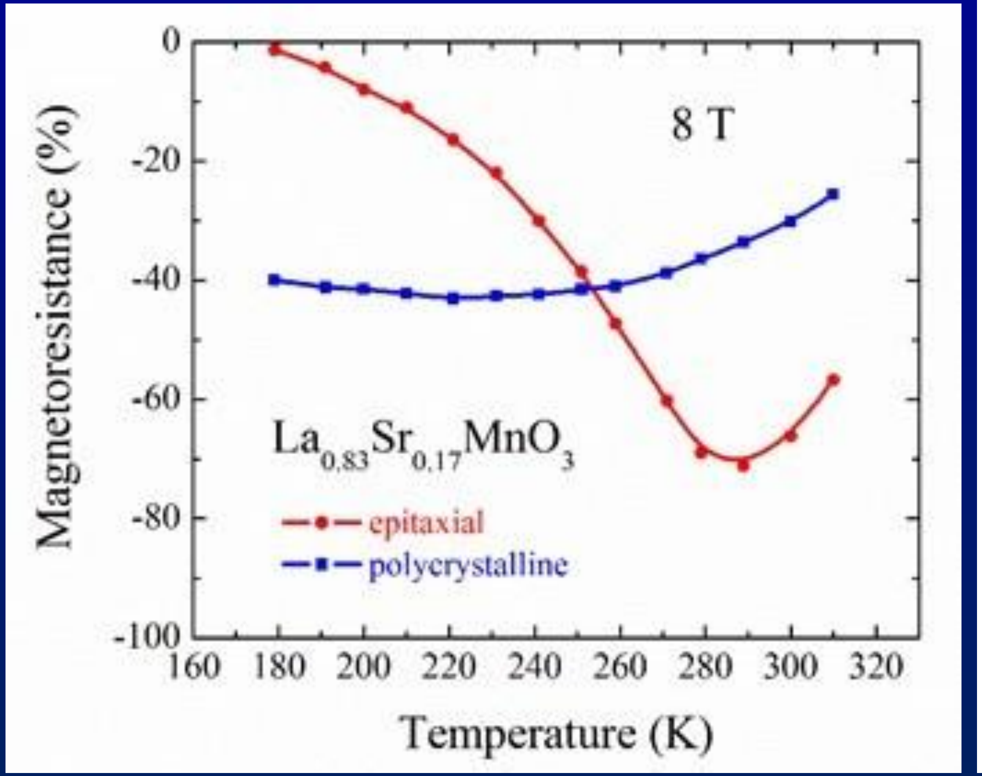
$La_{1-x}Ca_xMnO_3$

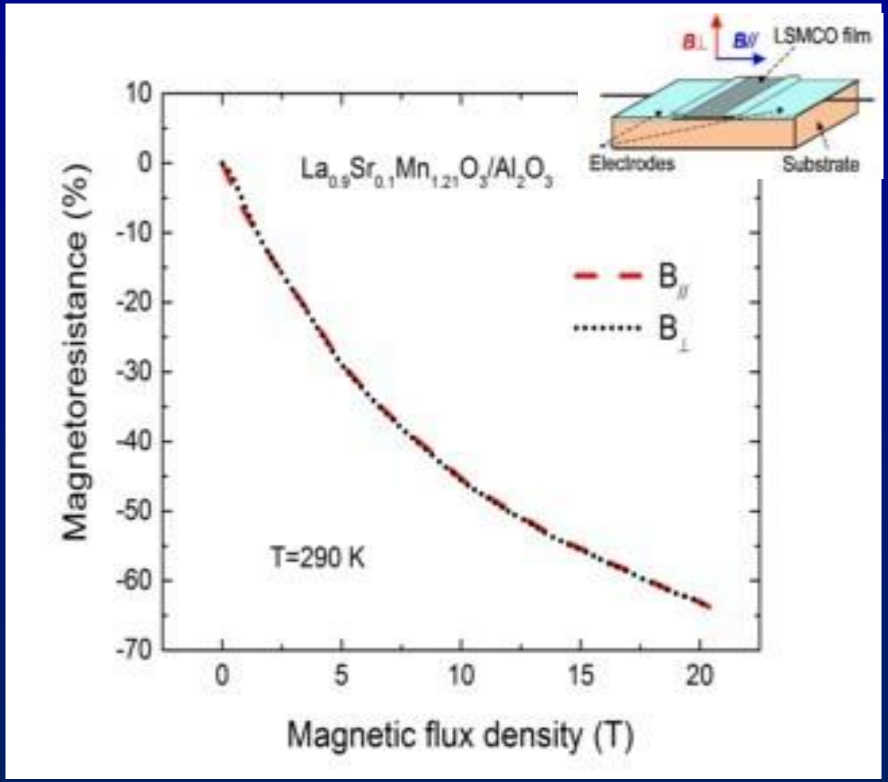
$La_{1-x}Sr_xMn_{1-y}Co_yO_3$

Why nanostructured (polycrystalline) films?









<u>Polycrystalline nanostructured manganite films are preferable for magnetic field sensors applications:</u>

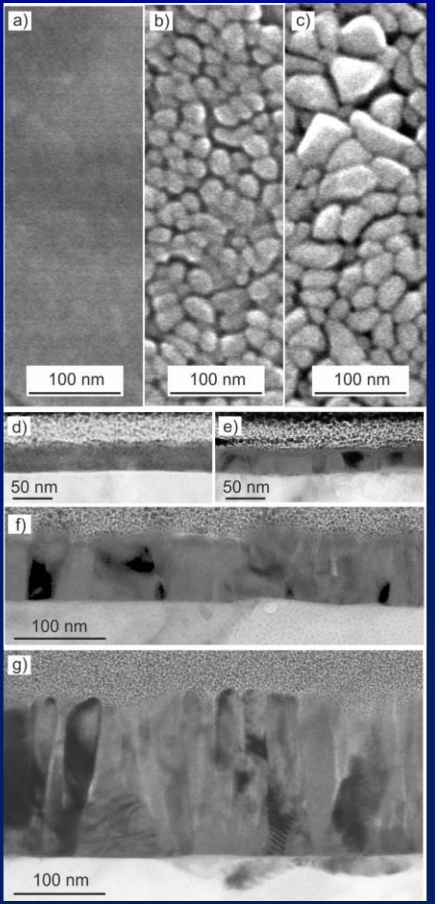
- ✓ **Sensitivity to magnetic field in a wide temperature range**
- ✓ **no saturation of MR up to very high fields**
- ✓ **Almost independent *MR* on field orientation** **CMR-B-scalar sensors**

Rev. Sci. Instrum., 85, 044704, 2014.

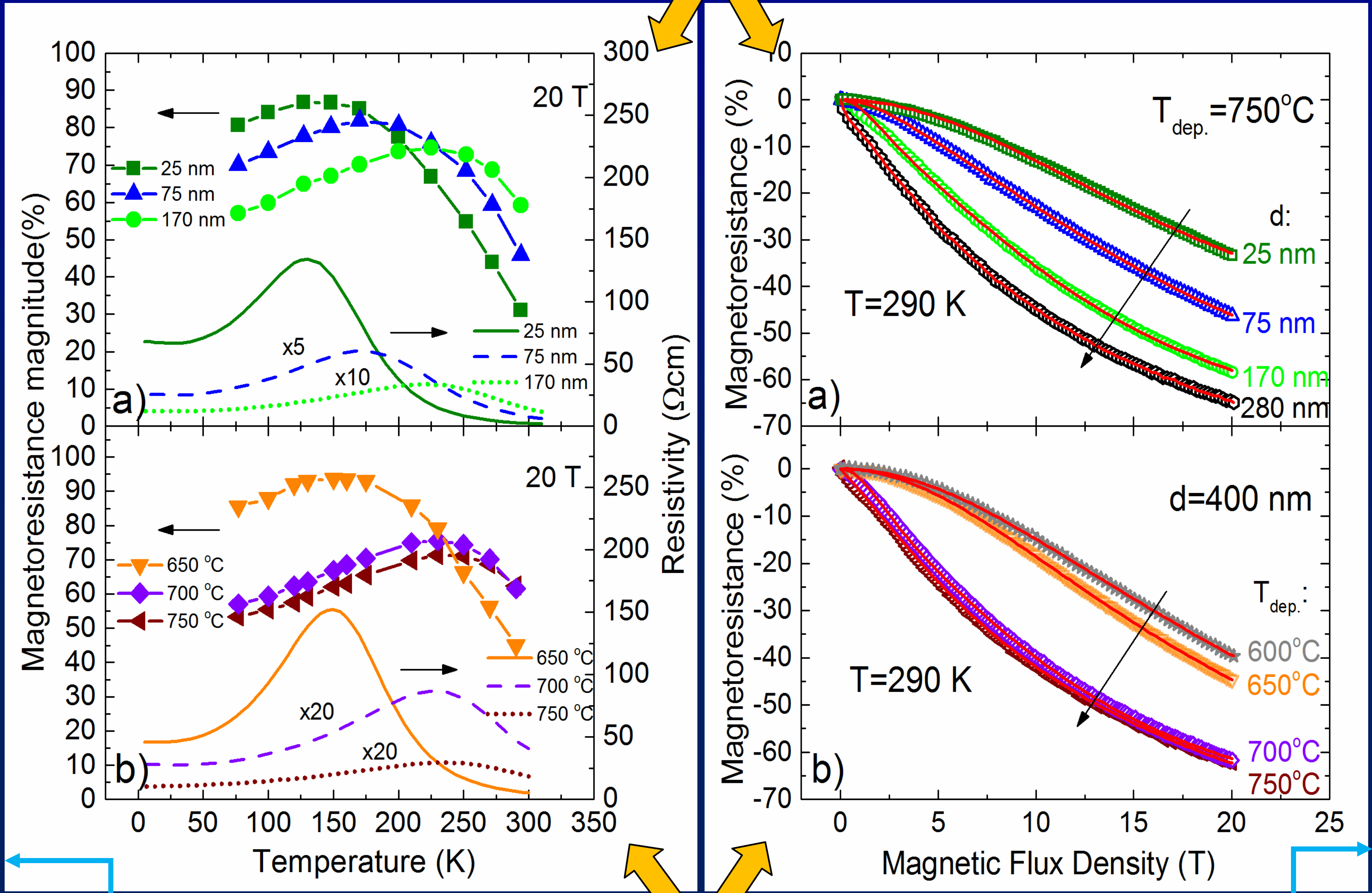
Magnetoresistance and resistivity

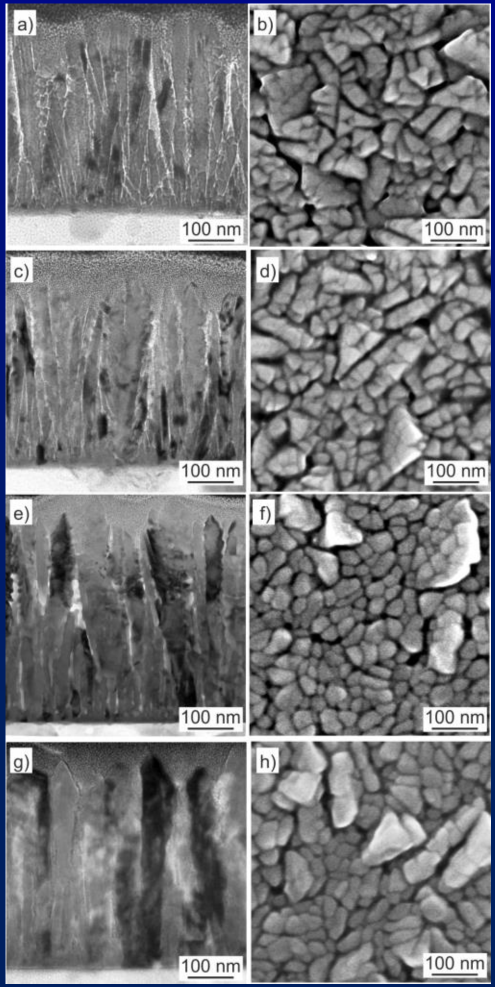
$La_{1-x}Sr_xMnO_3$ on lucalox substrate

dependence on film thickness



d=25 nm; 75 nm; 170 nm

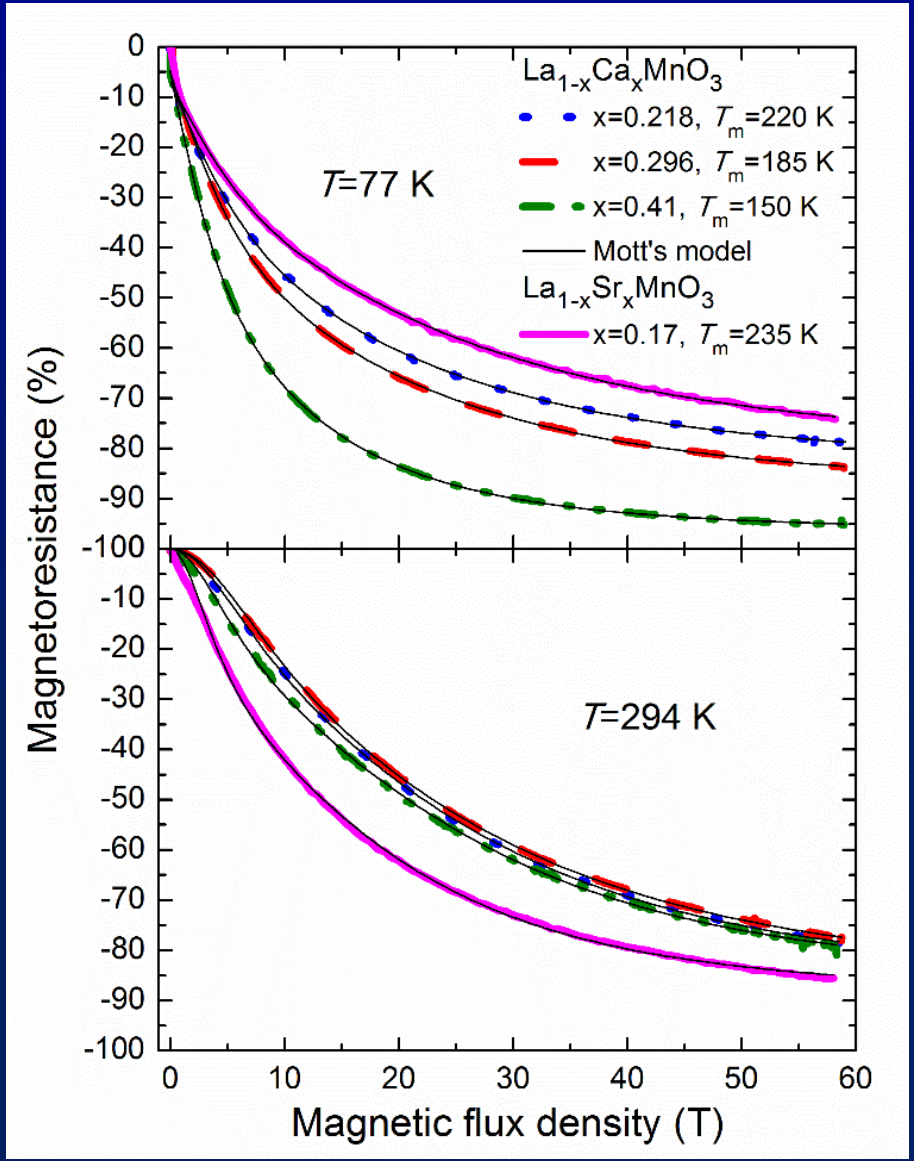


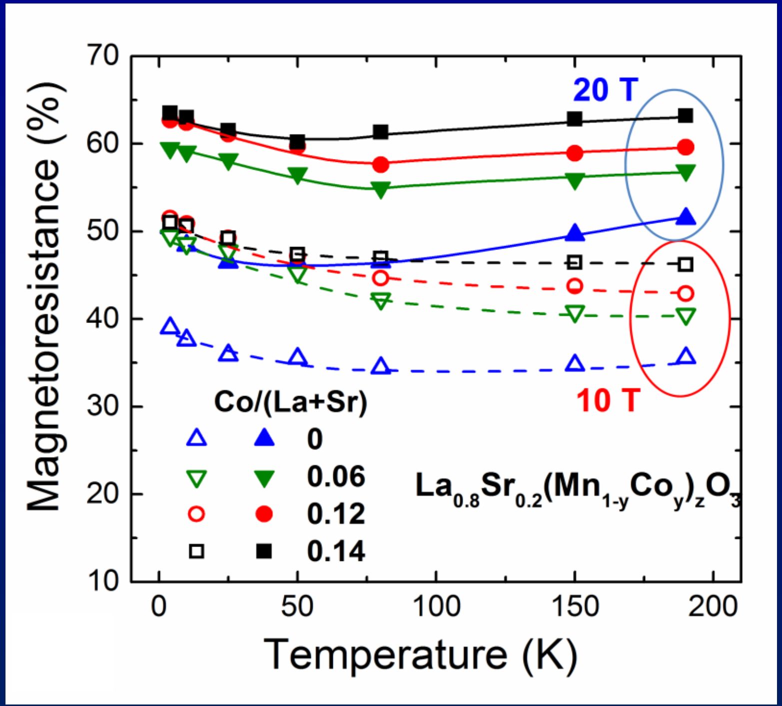


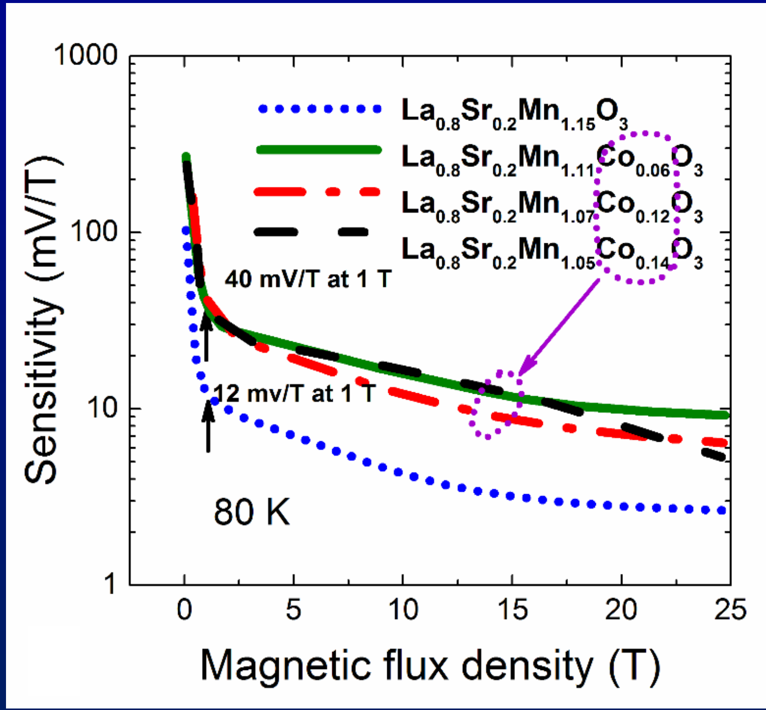
T_{dep}=600°C; 650°C; 700°C; 750°C

dependence on film deposition temperature

Comparison of magnetoresistance in $La_{1-x}Sr_xMnO_3$, $La_{1-x}Ca_xMnO_3$ and $La_{1-x}Sr_xMn_{1-y}Co_yO_3$ manganites







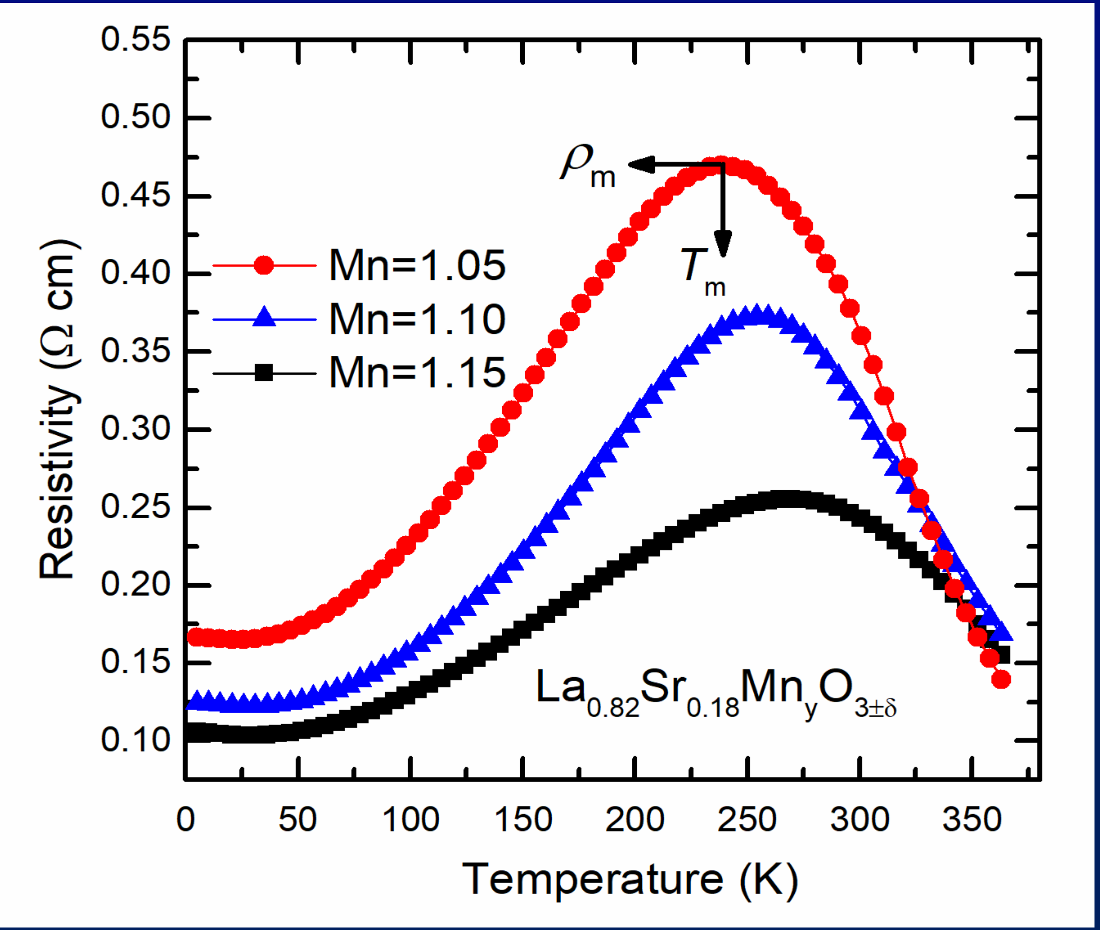
For pulsed magnetic field sensors operating at **cryogenic** temperatures **$La_{1-x}Ca_xMnO_3$** or **$La_{1-x}Sr_xMn_{1-y}Co_yO_3$** films which exhibit higher magnetoresistance values are preferable.

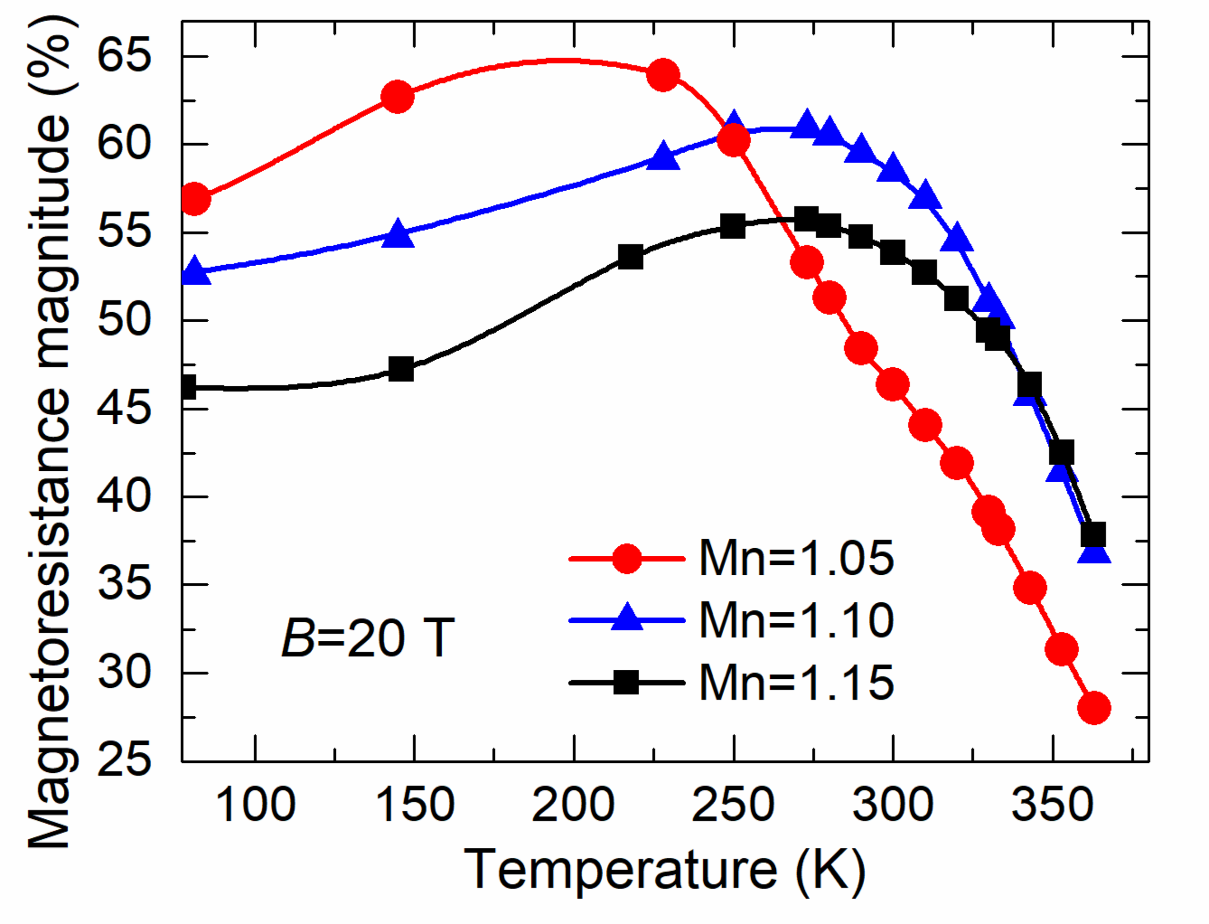
In comparison, **$La_{1-x}Sr_xMnO_3$** films having a higher sensitivity at room temperature should be favored for application at **high temperatures**.

Increase of operating temperature of magnetic field sensors based on La–Sr–Mn–O films with Mn excess

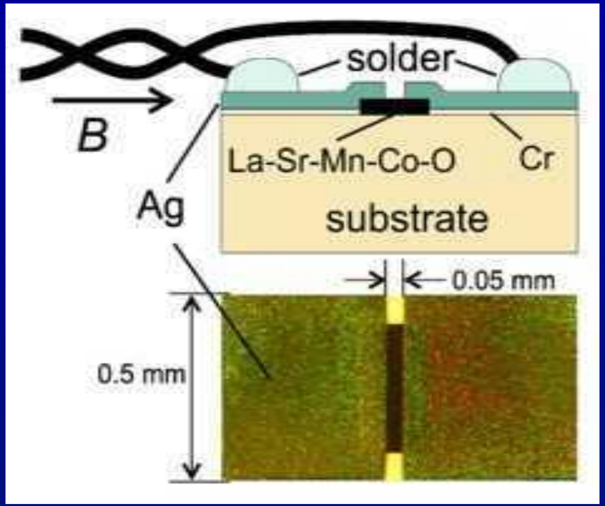
$La_{1-x}Sr_xMn_yO_{3\pm\delta}$ on polycrystalline Al_2O_3 substrate

Mn/(La+Sr)= 1.05; 1.10; 1.15





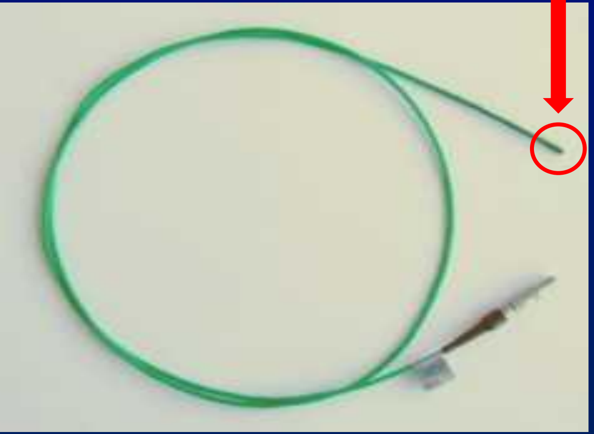
CMR-B-Scalar Magnetic field sensors: design

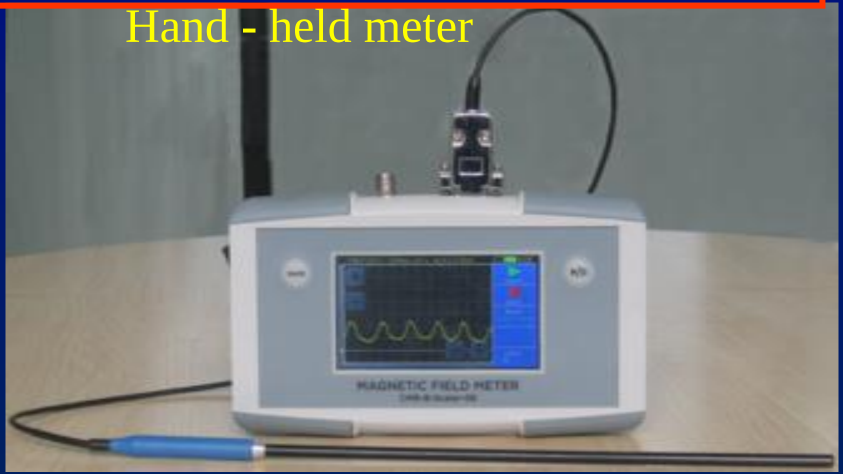


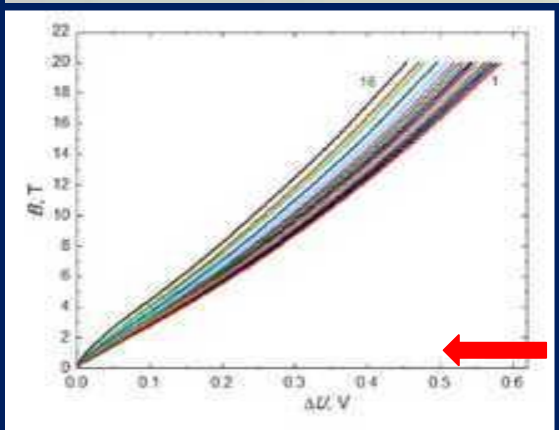
Magnetic field sensors were fabricated using a conventional integrated circuit processing technique. Electrical contacts were prepared by thermal deposition of silver (Ag) and separated by a distance of 50 µm. The contacts were annealed in argon atmosphere at 450 °C for 1h. The active volume of each sensor was 400 µm x 50 µm x 0.4 µm.

CMR-B-scalar meters (sampling rate 1-25 MS/s; resolution 40 µV; probe calibrated up to 30 T, temperature range 4-363 K)

CMR-B-scalar sensor

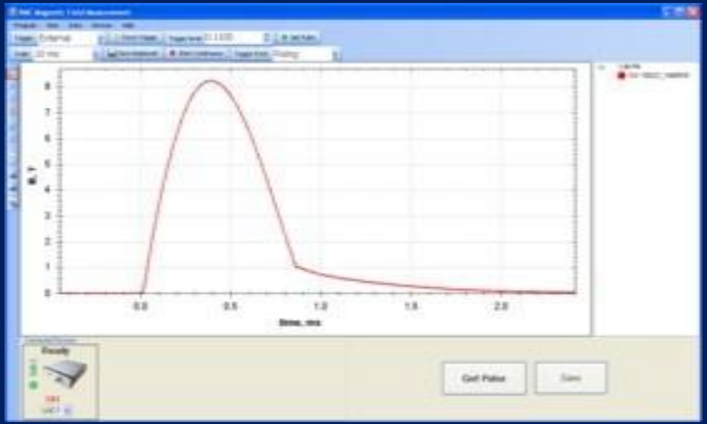






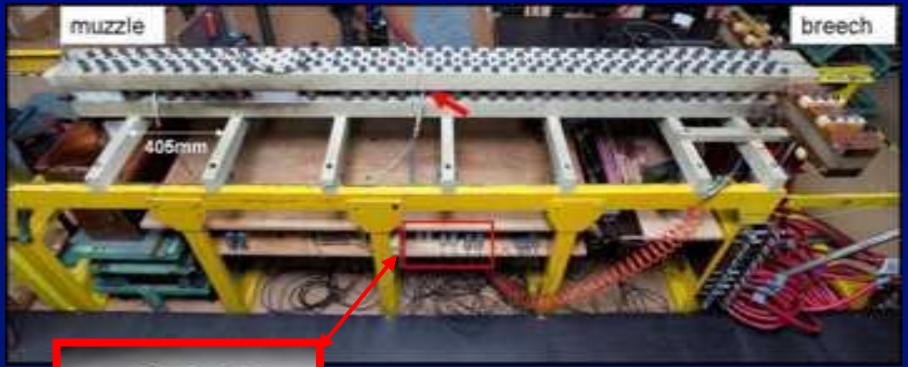
Calibration curves of CMR-B-scalar sensor: 1 - 0°C, 16 - 45°C

Measured magnetic field pulse

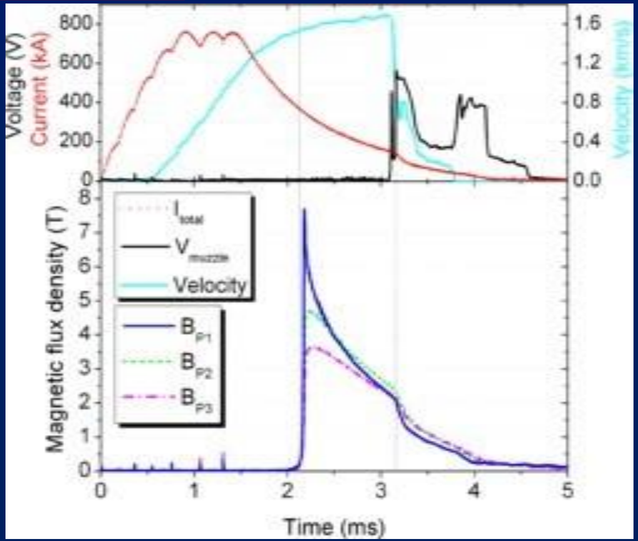


Application of La-Sr-Mn-O Films: CMR-B-Scalar Sensors

Velocity-Induced Current Profiles Inside the Rails of an Electromagnetic Launcher

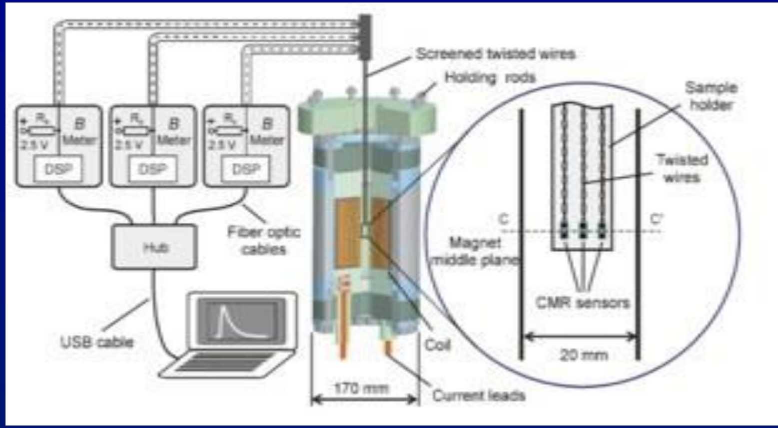


Measurement signal at ISL railgun RAFIRA shot with a projectile velocity of 1500 m/s

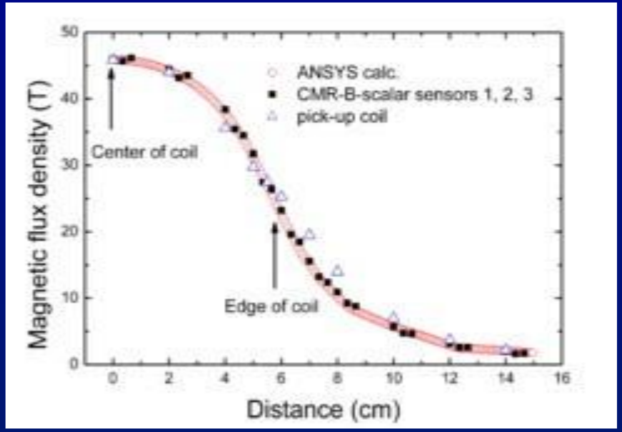


IEEE Trans. Plasma Science, v.41, p.1520, 2013

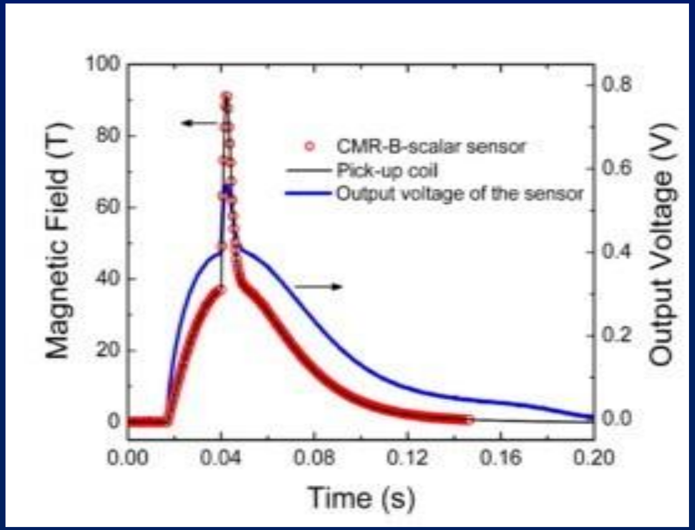
High Magnetic Field Measurement in Non-destructive Pulsed Magnets



Axial magnetic field measurements



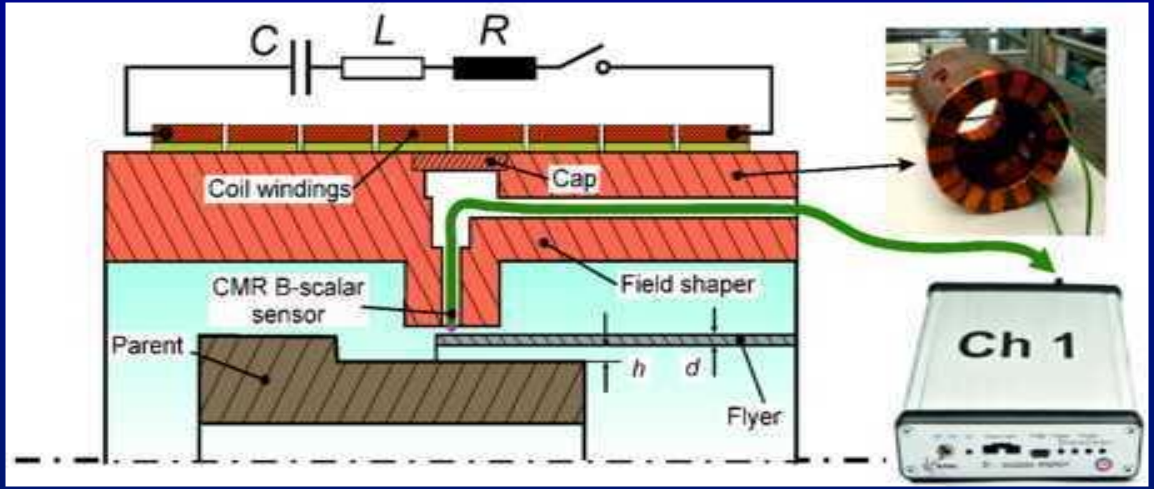
IEEE Trans. Magnetics, 49, 5480, 2013



Appl. Phys. Lett., 101, 092407, 2012

On June 22, 2011, the Helmholtz-Zentrum Dresden-Rossendorf set a World record for the strongest magnetic field ever delivered by a nondestructive magnet: 91.4 Tesla; 94.2 Tesla on Jan 2012.
Los Alamos National Laboratory (LANL) 100.75 T on March 22, 2012

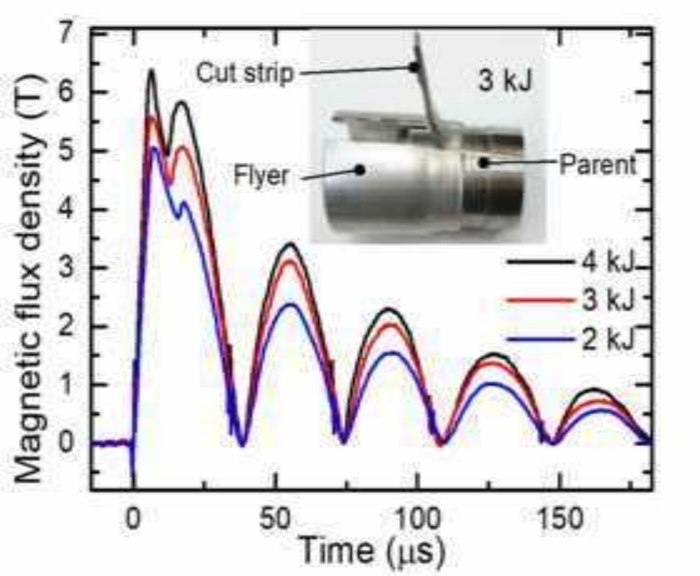
Magnetic field measurements during magnetic pulse welding (MPW)

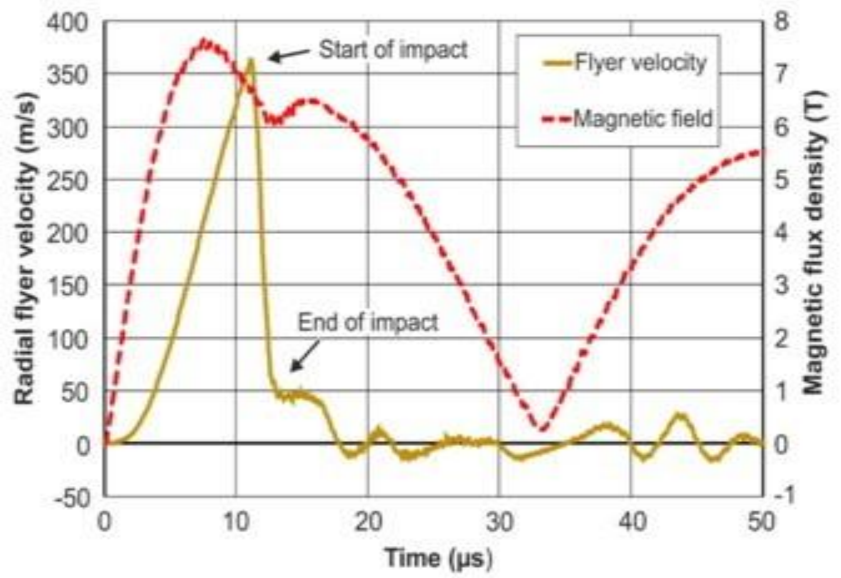


Schematic diagram of setup installed at Institute of Forming Technology and Lightweight Components, TU Dortmund University

Magnetic field measured between the field shaper and aluminum flyer tube:

Possibility of nondestructive welding quality control by analysis of magnetic pulse shape.





LS-DYNA simulation

V. Stankevic et al. Sensors 2020, 20, 5925

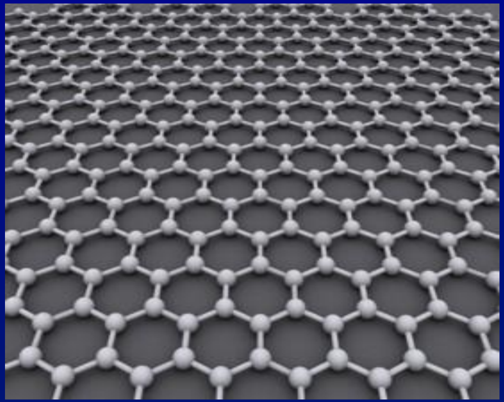
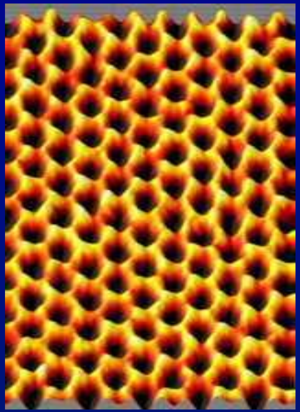
CMR materials – summary

- **Ferromagnetic oxide materials (Lanthanum manganites)** exhibiting **Colossal magnetoresistance (CMR) effect**. Due to reorientation of magnetic moments of atoms (for example, manganese in La-Sr-Mn-O) in applied magnetic field the resistivity of material decreases: **negative magnetoresistance. High sensitivity in low and intermediate magnetic fields, however, saturates at high fields.**

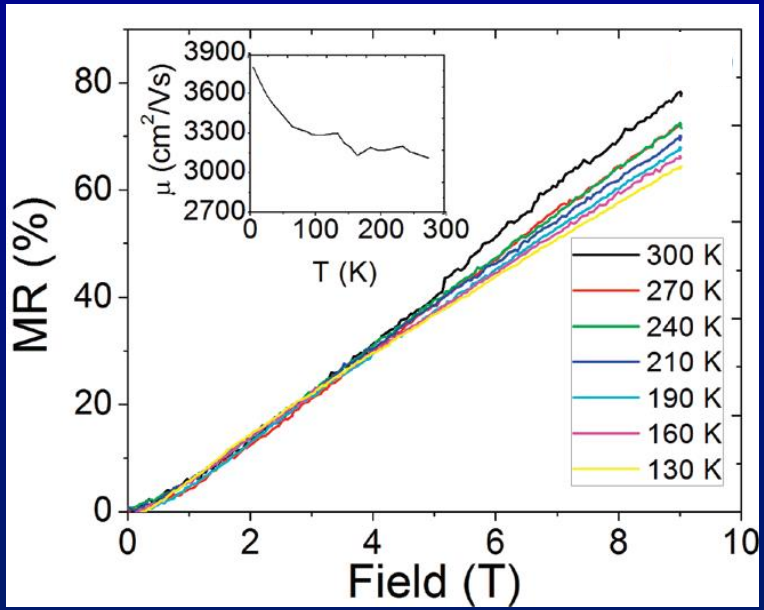
2D materials

- **Two-dimensional (2D) semiconductors (graphene) – Lorentz force induced positive magnetoresistance (Gauss effect).** High and linear magnetoresistance at high magnetic fields, however, small in low-field limit.

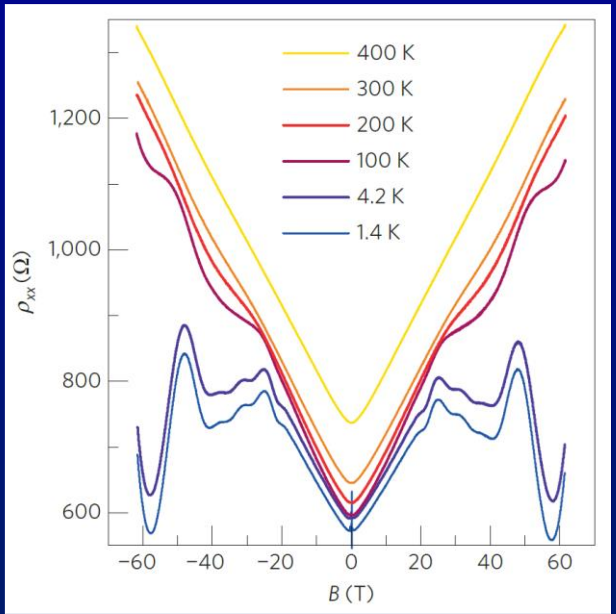
2D materials: graphene

a  b 

Graphene is an atomic-scale hexagonal lattice made of carbon atoms:
a) schematic representation; b) STM image.



Friedman, *Nano Lett.* **10**, 3962, 2010



Kisslinger, *Nature physics* **11**, 650, 2015

Magnetoresistance reveals **a quadratic dependence** on magnetic field **in the low-field limit.** Semiclassically, the strong-field magnetoresistance saturates quickly to constant value C.

$$\frac{\Delta\rho}{\rho} \propto \begin{cases} (\mu H)^2, & \mu H < 1 \\ C, & \mu H > 1, \end{cases}$$

where μ is the carriers' mobility

At still higher magnetic fields, it is possible to reach the 'extreme quantum limit' (Abrikosov):

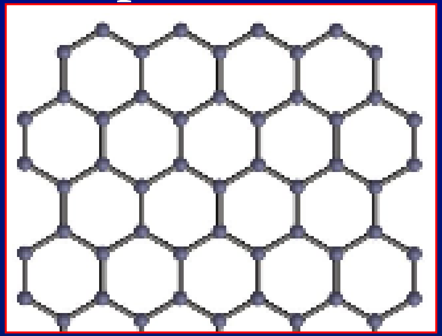
$$\rho_{xx} = \rho_{yy} = \frac{N_i H}{\pi n^2 ec} \propto H$$

where n is the density of electrons and N_i is the concentration of static scattering centres

Graphene/manganite structure: application for magnetic field sensors

Main idea:

Graphene SLG



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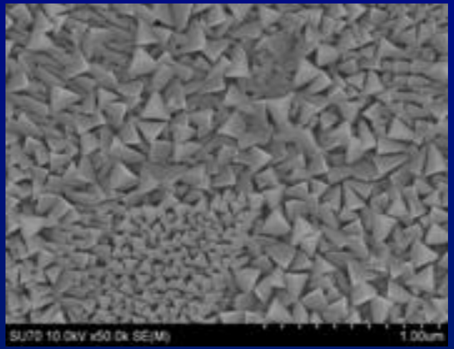
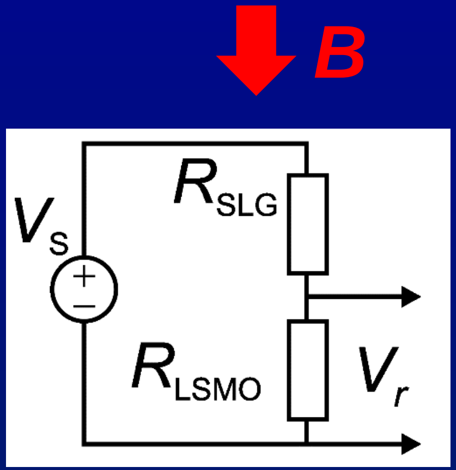
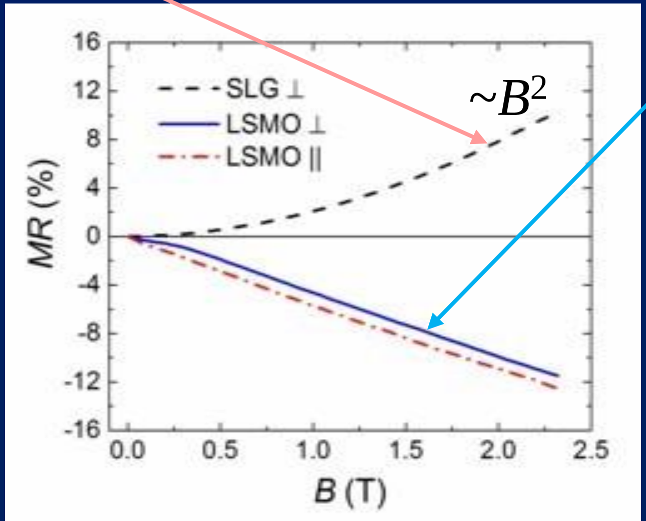
Manganite LSMO





Positive magnetoresistance in graphene (Lorentz force)

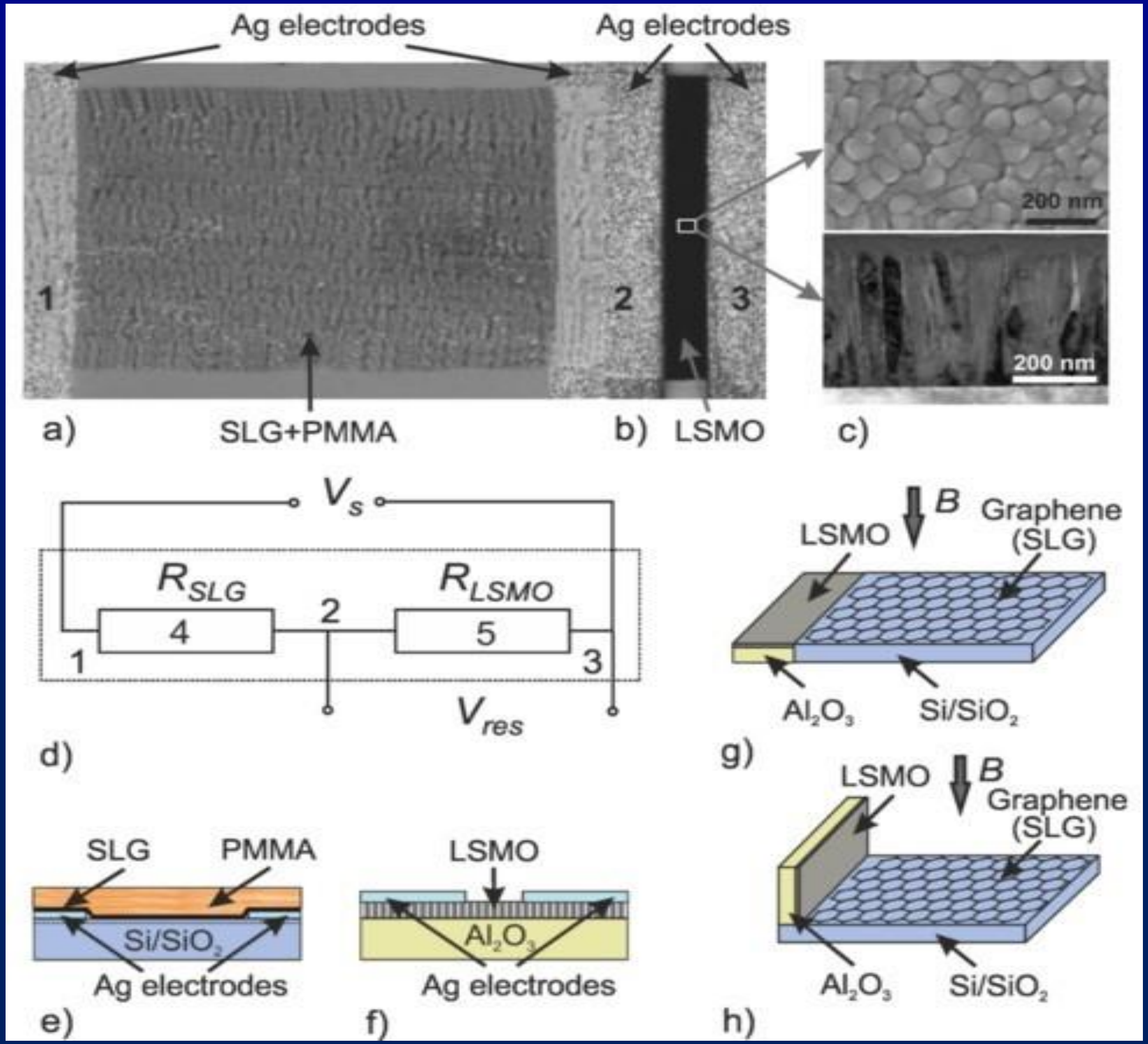
Negative magnetoresistance in manganite (CMR effect)



Hybrid sensor:
higher response signal?

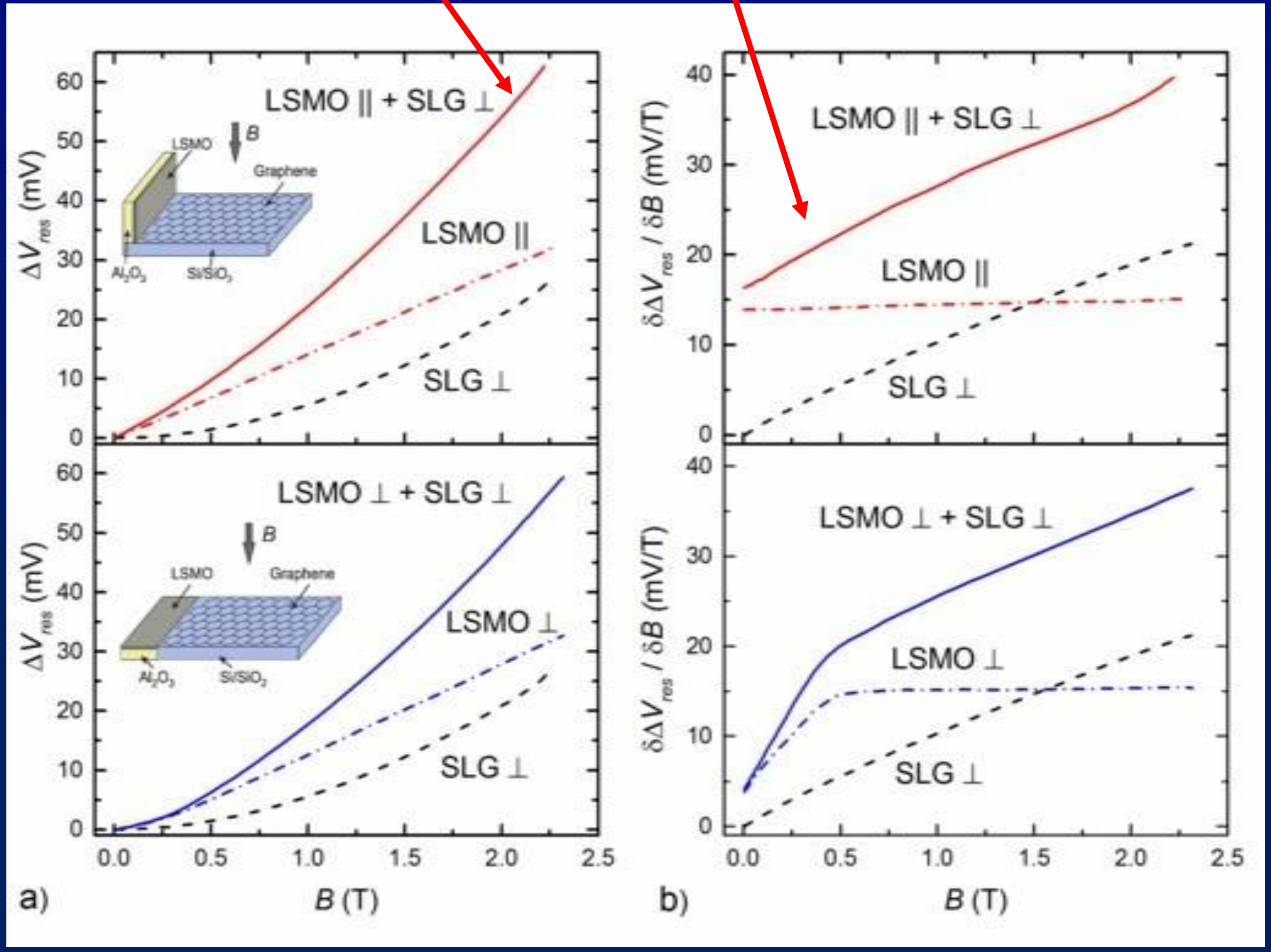
Hybrid Graphene/Manganite Sensor

Realization



Testing and validation

Sensor's **response** and **sensitivity** increases several times in comparison with single graphene



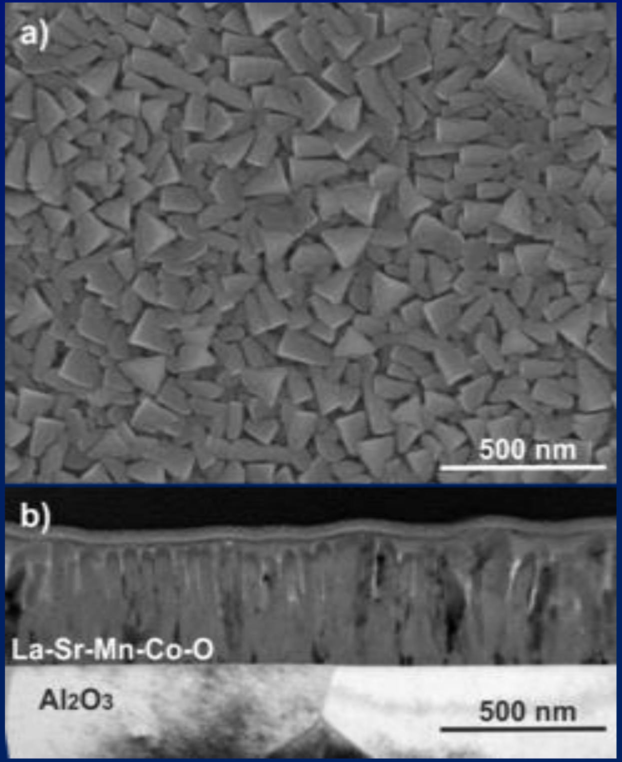
R. Lukose et al. ***Nanotechnology*** 30 (2019) 355503

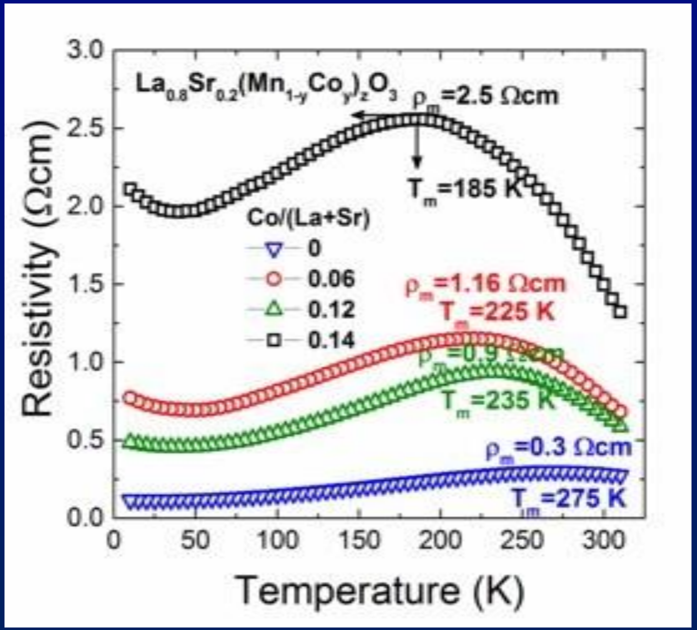
Hybrid Graphene/Manganite Sensor:
Increase of manganite sensitivity

Co-substituted manganite film

$La_{1-x}Sr_xMn_{1-y}Co_yO_3$

Nanostructured films

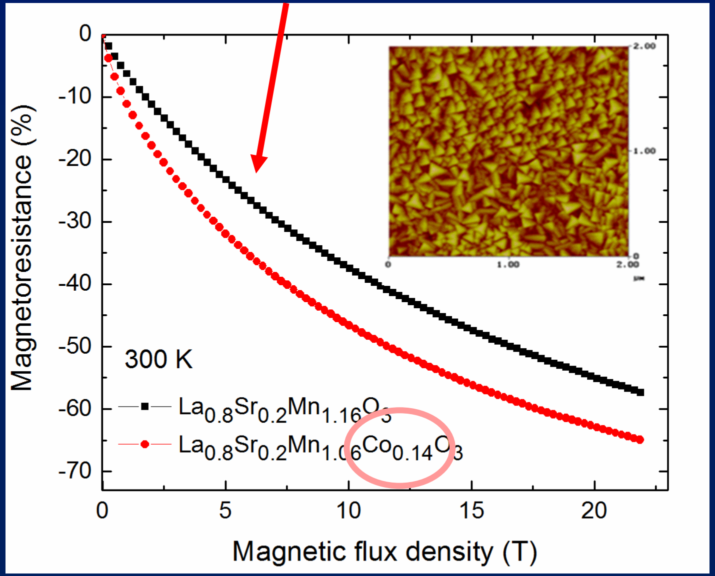




Co substitution partly destroys double exchange mechanism:

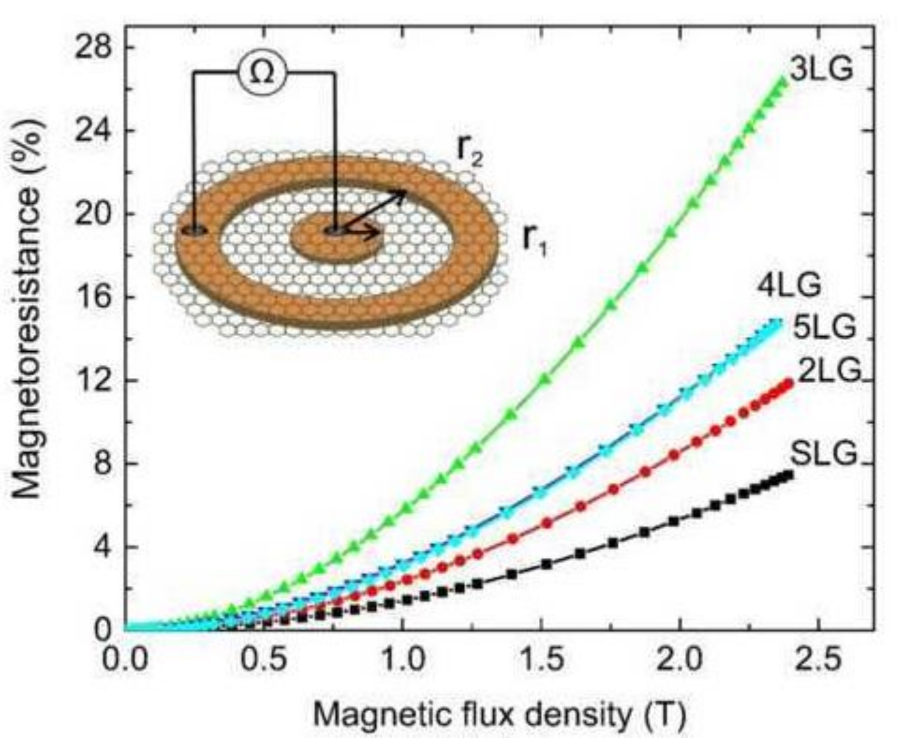
Mn^{3+}–O–Mn^{4+}

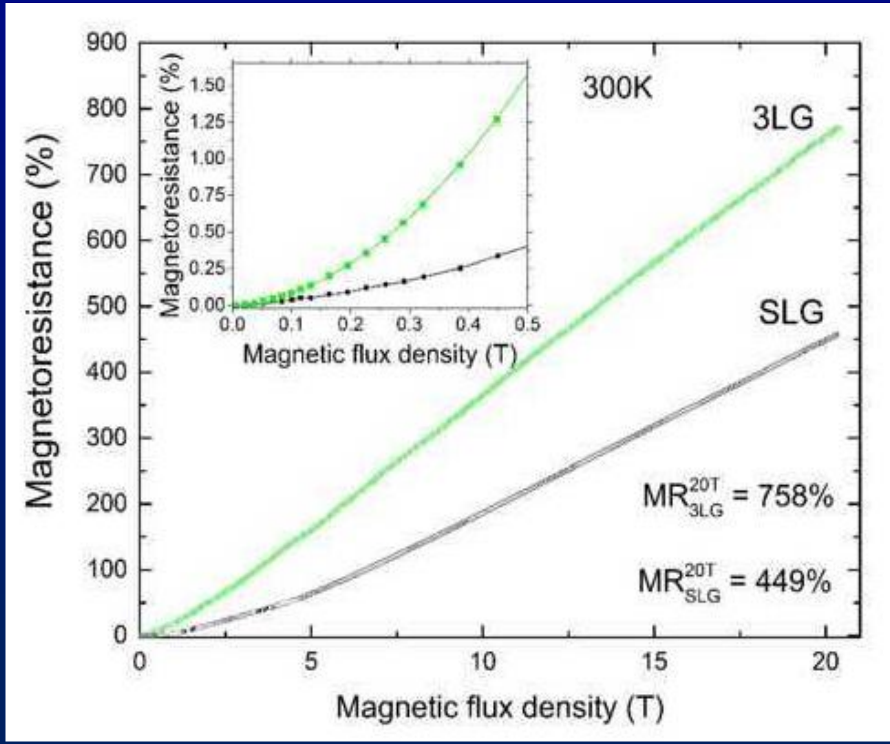
Increase of Co amount increases magnetoresistance!

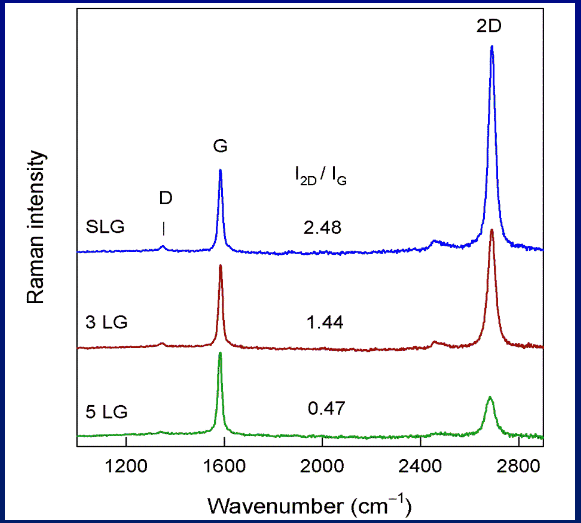


Hybrid Graphene/Manganite Sensor: increase of graphene response to magnetic field

Corbino disc configuration. Number of graphene layers: n=1,2,3,4,5







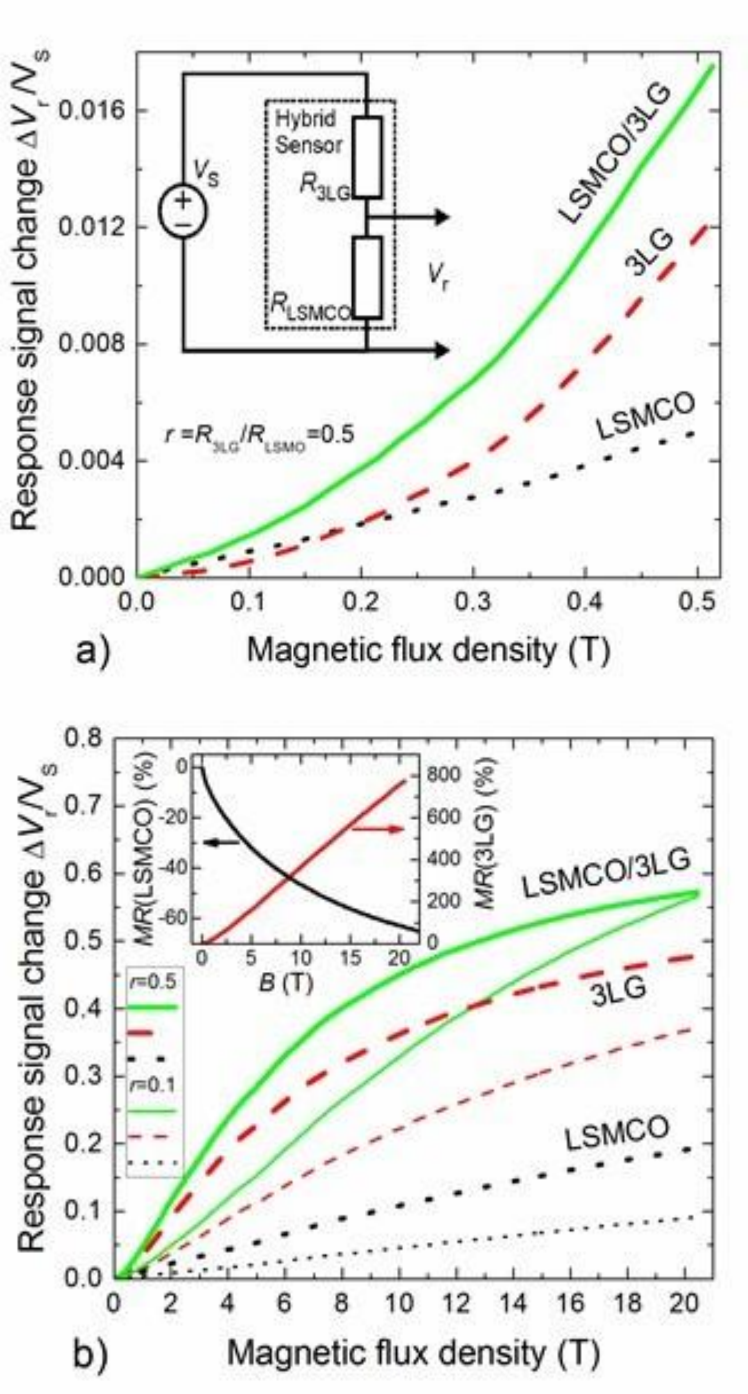
The Raman intensity ratio of the 2D band to G (I_{2D}/I_G) was used to probe the number of graphene layers

Mobility (μ) evaluated from dependence $MR \sim (\mu B)^2$:
SLG 1180 cm^2/Vs;
2LG 1530 cm^2/Vs;
3LG 2420 cm^2/Vs;
4LG 1790 cm^2/Vs;
5LG 1780 cm^2/Vs.

For comparison: Liao *et al. (Adv. Mater. 24, 1862, 2012)* μ≈540 cm^2/Vs at 2 K and ≈310 cm^2/Vs, at T= 300 K

Increased magnetoresistance of 3LG !

Lukose *et al. Scientific Reports* 9, 9497, 2019



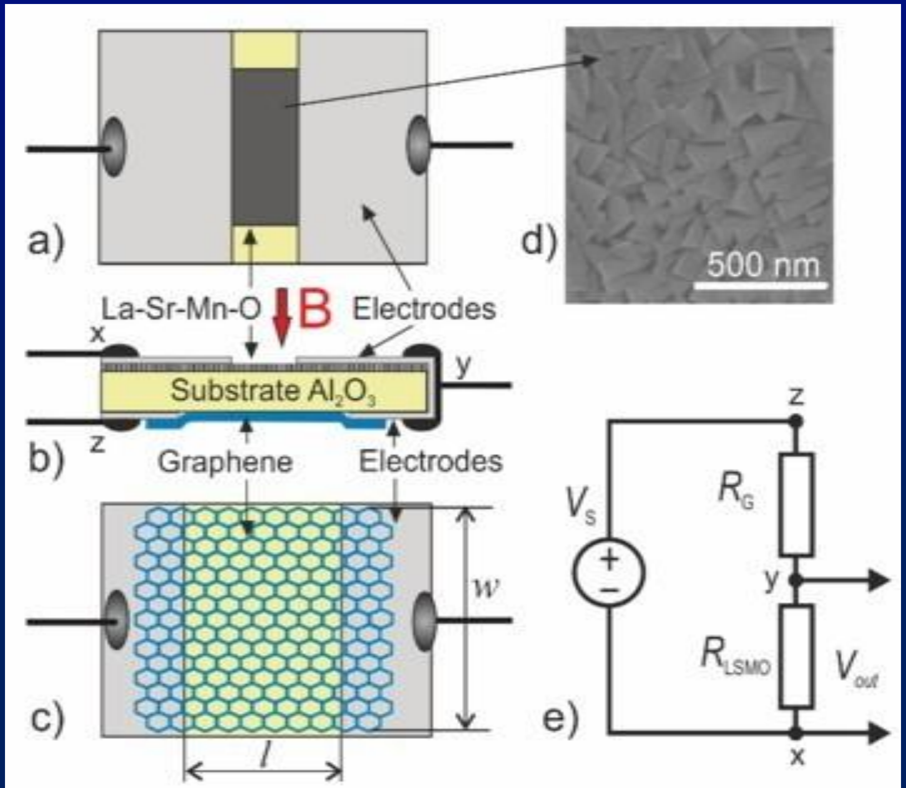
Hybrid manganite-cobaltite/3LG sensor

Sensor	Sensitivity S, mV/(V*T) (at B=0.5T)	S maximal, mV/(V*T) (field range B)	S, mV/(V*T) (at B=20 T)
LSMCO **$R_{bal}/R_{LSMCO}(0)=0.5$**	12	12 (B=0.3÷10 T)	7
3LG **$R_{3LG}(0)/R_{bal}=0.5$**	40	56 (B=1÷2 T)	7
Hybrid **$R_{3LG}(0)/R_{LSMCO}(0)=0.5$**	**57**	**70** **(B=1÷2 T)**	**7**
LSMCO **$R_{bal}/R_{LSMCO}(0)=0.1$**	5	5 (B=0.3÷20 T)	5
3LG **$R_{3LG}(0)/R_{bal}=0.1$**	17	27 (B=2.5÷7 T)	10
Hybrid **$R_{3LG}(0)/R_{LSMCO}(0)=0.1$**	**21**	**40** **(B=2.5÷7 T)**	**17**

The sensitivity of the hybrid sensor could be increased by changing the ratio of zero-field resistances of manganite-cobaltite and graphene

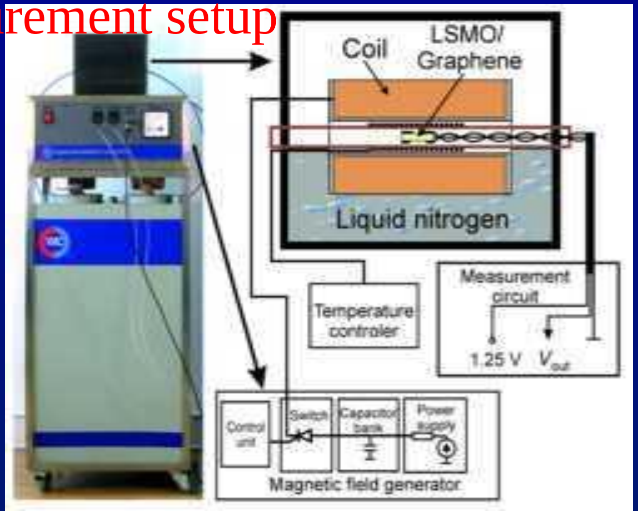
Compact magnetoresistive manganite/3L graphene sensor

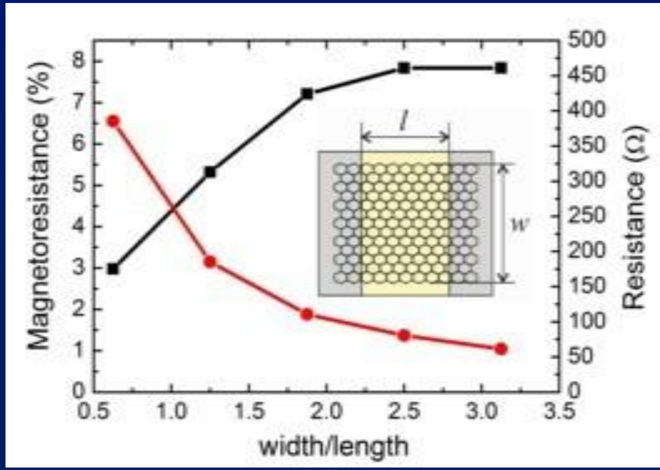
For local magnetic field measurements:
prepared on both sides of the same Al_2O_3 substrate



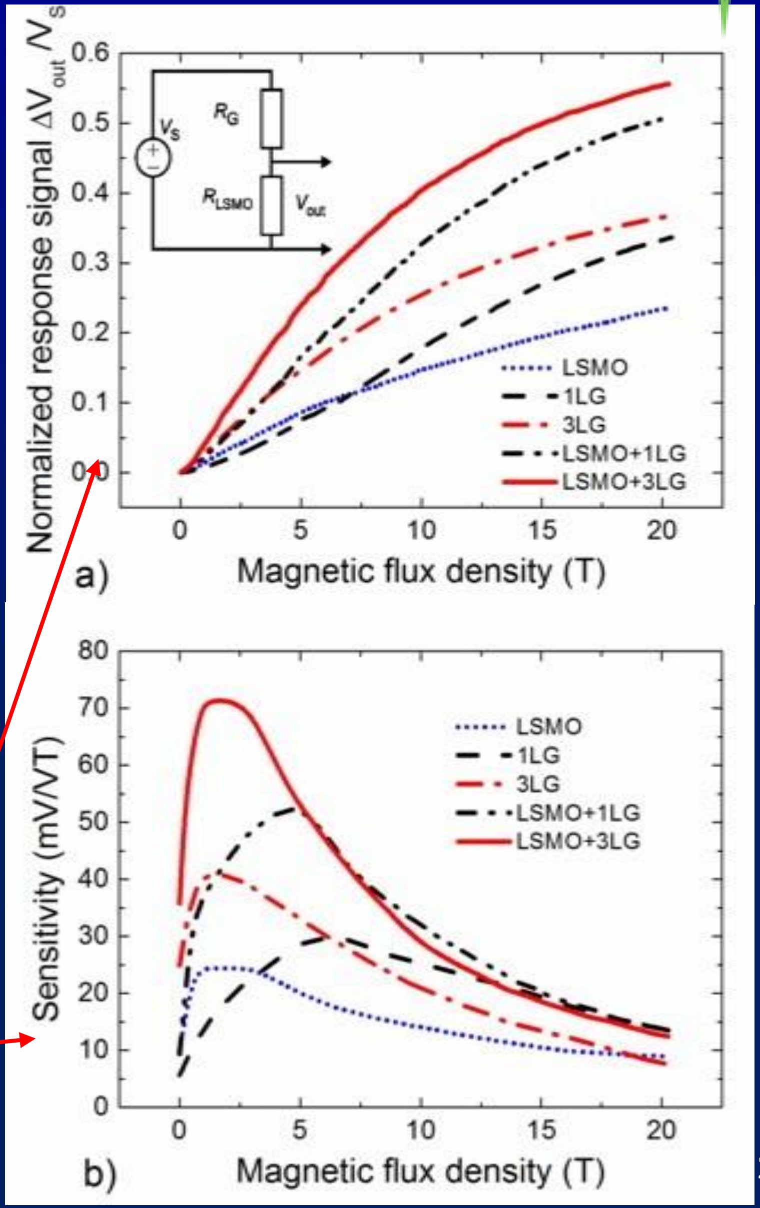
w/*l*=3, *l*=200 μm

Measurement setup



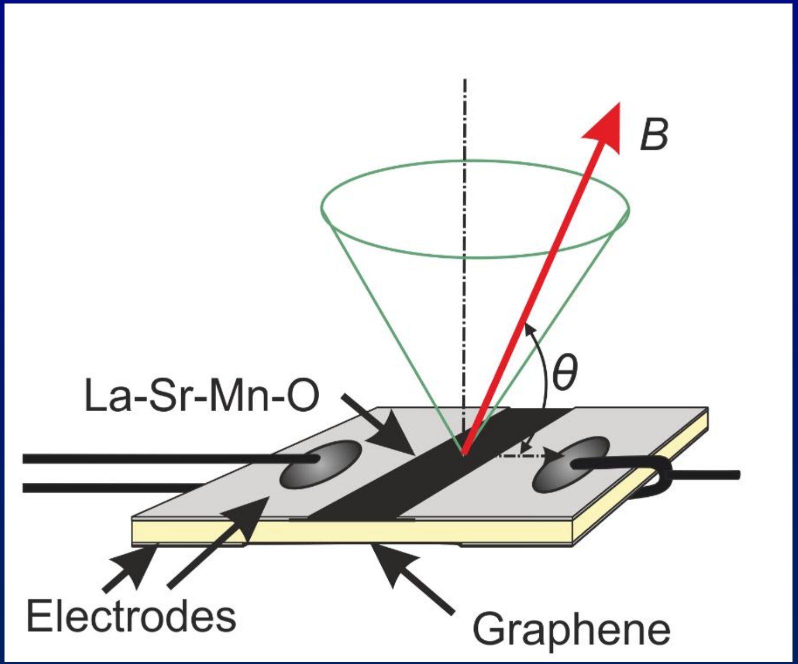


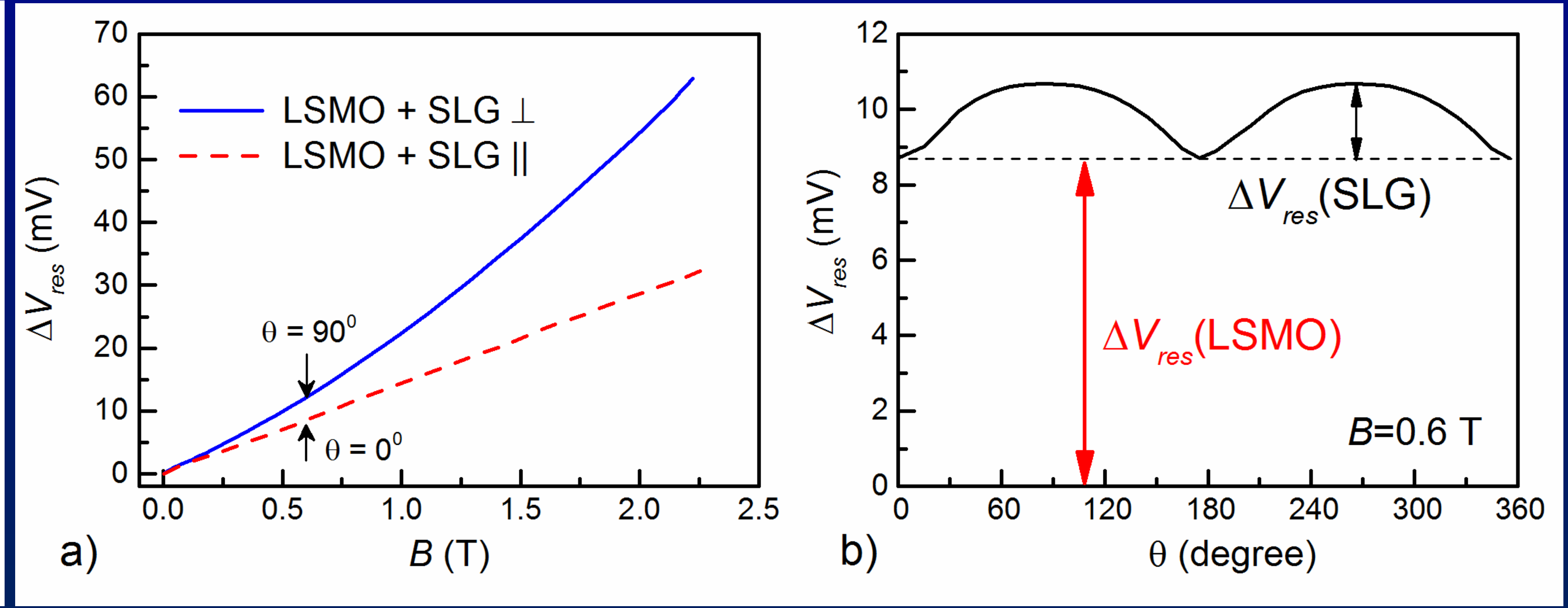
Increased response signal and sensitivity in a wide range of magnetic fields



Zurauskiene *et.al*, *IEEE Magn. Lett.* 10, 8105605, 2019

Measurement of absolute magnetic field value and direction in respect to sensor's plane





Schematic representation of hybrid manganite-graphene sensor and magnetic field direction

Response signal of hybrid manganite-graphene sensor

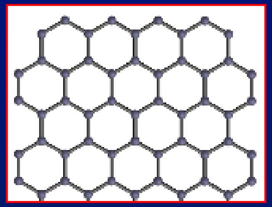
Response signal of hybrid manganite-graphene sensor at different magnetic field angles θ in respect to the graphene plane

Summary and future perspectives

CMR-B-scalar sensors
(based on nanostructured manganite films)

Specific industrial and scientific applications:
electromagnetic launching; magnetic forming and welding; investigations of synchronous and linear motors; development of pulsed field magnets, etc.

Hybrid graphene/manganite sensors



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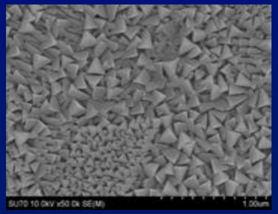
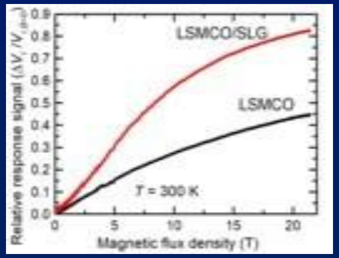
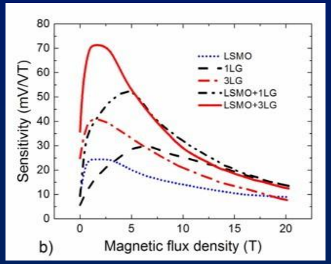


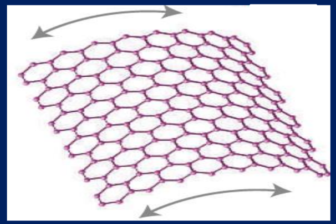
For local measurements with increased response signal and sensitivity in a wide magnetic field range.





Future prototypes:

- measurement of magnitude and field direction in respect to sensor’s plane;
- further increase of response signal and sensitivity;
- scaling dimensions to micrometer size;
- application in wide range of temperatures;
- flexible hybrid sensors?



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Thank you for your attention !

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