Plasmonic approaches for spin-polarized metallic photocathodes

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Abstract

- We have explored new photocathode materials and schemes to develop strategies and technologies for next generation nuclear physics accelerator capabilities.
- We have applied an experimental setup for light incidence at an acute angle onto the photocathode surface in UHV, in order to excite surface Plasmon resonance, hence increasing light absorption by the metallic surface and test the photo-emitted current. We have also used oblique incidence thin film deposition onto gratings to achieve optimized Plasmonic excitation leading to stronger EM field and also lower emittance.
- In parallel, we investigated magnetic thin films incorporating plasmonic materials using alloys such as “Silmanal”. We have been able to achieve such films and are at present investigating deposition onto gratings to extend our studies onto spin-polarized photoemission.

Next-generation photocathode requirements

- Photocathodes are critical components of photo-injectors, where the photocathode must produce an electron beam with stringent requirements on emittance (ε, i.e. the average spread of particle coordinates in position-and-momentum phase space), temporal response, life-time and in some cases polarization to match the properties needed for a particular application.
- Next-generation light sources will also need megahertz repetition rates, and since it's electrons that make the x-rays, the only way to achieve that kind of performance is with an electron gun that can deliver tight electron bunches with high charge, high energy, and a very high repetition rate. In addition, the next generation electron ion colliders (EIC) will require at least 50mA of polarized e-beam in order to achieve the desired luminosity at the electron/proton interaction region.
- At present, strained-layer superlattice GaAs/GaAsP photocathodes represent the state of the art for generating polarized beams with polarization approaching 90% and photoelectron yield or quantum efficiency about 1%.

The problem

- Maximizing brightness requires increasing the pulse charge via increasing QE and reducing ε. However, both of these quantities depend on the electron excess energy: $\hbar\omega - \phi_{eff}$

$$\epsilon = \sigma_x \sqrt{\frac{\hbar\omega - \phi_{eff}}{3mc^2}},$$

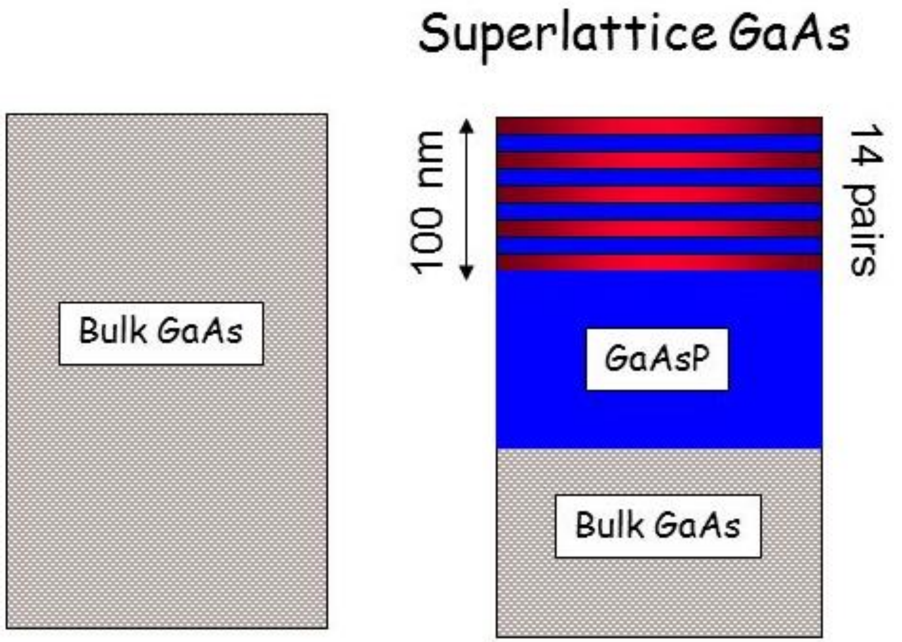
$$QE \approx \eta_{tr} \frac{\left(\hbar\omega - \phi_{eff}\right)^2}{8\phi_{eff}\left(E_F + \phi_{eff}\right)},$$

- where ϕeff is the metal’s work function, EF is the metals Fermi level, σx is the beam size, and ηtr is the probability that a photon is absorbed and transported to the surface without scattering. The minimum beam size σx is given by the space charge limit so the only way to reduce ε is to reduce the excess energy, which, in turn decreases metal’s QE reducing the pulse charge.

Goal: to attempt more robust metallic photocathodes

- **For this approach it is paramount to enhance the quantum efficiency (QE) of metallic-based photocathode. This can be achieved in principle by excitation of surface plasmon resonance(SPR)**
- **Present approach:**

QE (maximum) ~20% at 532nm; 11 hrs of lifetime at 100mA, 1000C



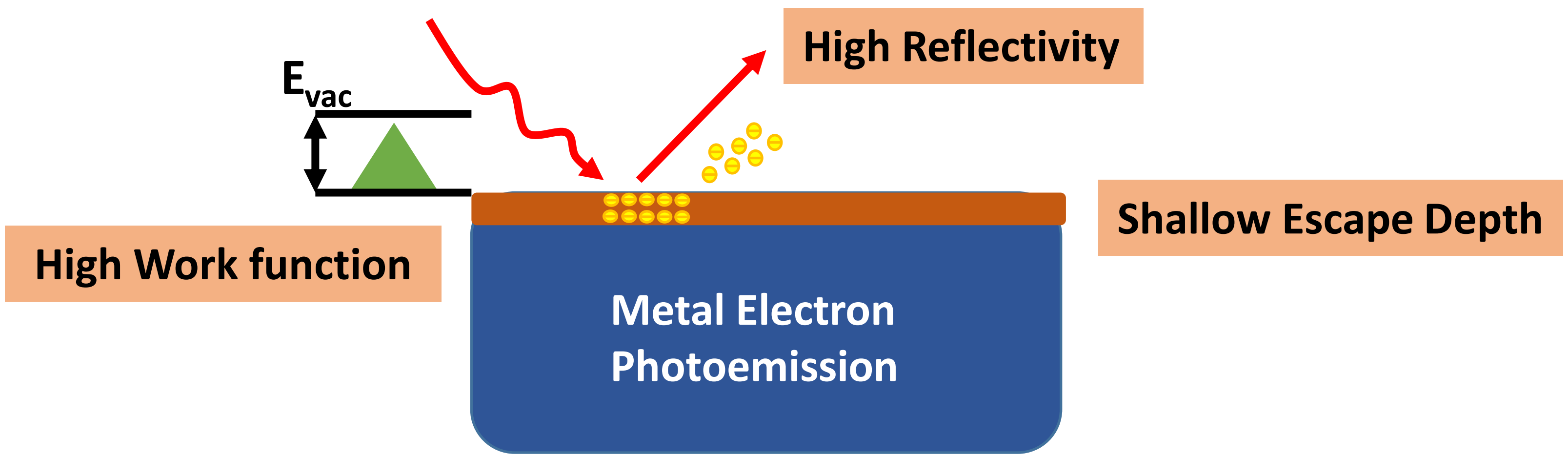
Polarization 80% ~90%; QE (maximum) ~1% at 780nm laser, 1hr of lifetime at 50mA, 1000C

Semiconductor vs metals

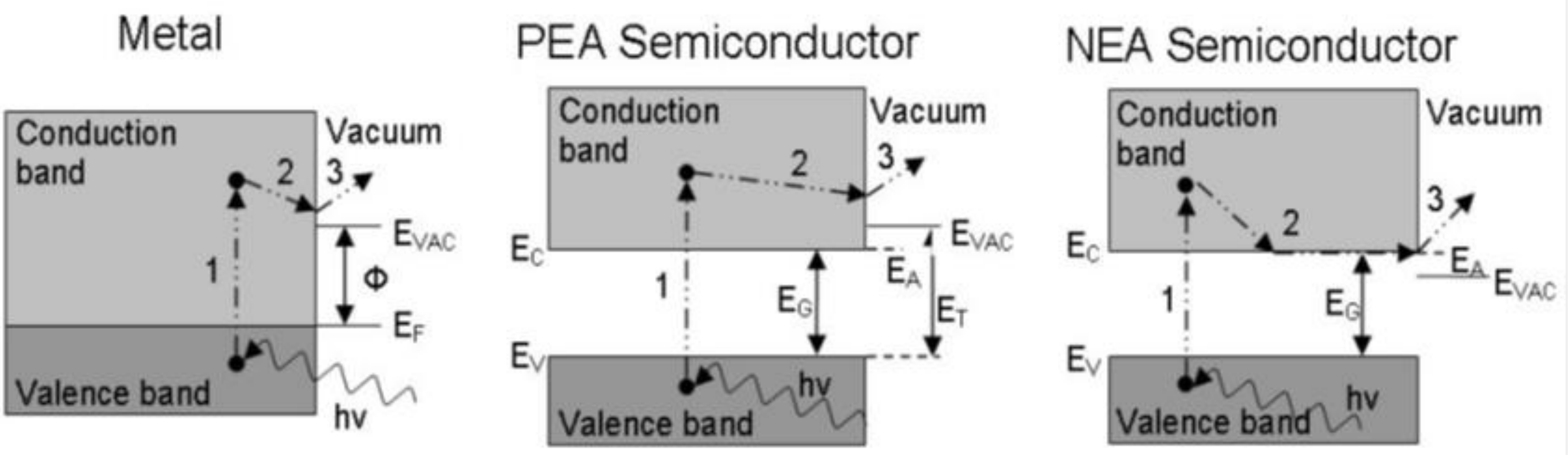
- Semiconductors have offered some advantages as photocathodes because when an electron is excited to the conduction band it does not scatter with other electrons since there are no states in the gap. A disadvantage is that an excited electron can stay for a long time inside the bulk after excitation before exiting the material, which produces a temporal spread from a semiconductor photocathode.
- In metals most of the excited electrons scatter with electrons in the bulk as they make their way to the surface, thus each scattering is catastrophic, i.e. the energy lost due to a single scattering event yields an electron with a below-work function total energy, which results in poor QE, but the temporal response is on the same time scale as the pulse length of an incident pulsed laser beam, because only those electron that travel directly to the surface without electron-electron scattering are able to exit the bulk material

Comparison between metallic and Semiconductor Photocathodes

Metal Photocathode	Semiconductor Photocathode
Low QE ~ 10^{-5} at 405nm for Ag	High QE ~20% at 532nm
Long life time ~ years	Short life time ~ hours or days
Short response time ~ ps	Longer response time ~ 10 ps
Lower requirement for vacuum	High requirement for vacuum ~ 10^{-10} Torr



Photoemission 3-step model: 1.Optical excitation of electrons from the valence band into higher empty states in the conduction band. 2. Part of these electrons migrates to the solid surface, either directly or by experiencing scattering with the lattice or other electrons. 3. Escape across the surface potential barrier into vacuum of those electrons that have sufficient energy left at the surface.



Sketch of the three-step model for photoemission of electrons for metals and semiconductors. PEA and NEA are positive and negative electron affinity semiconductors respectively

A technology that squeezes electromagnetic waves into minuscule structures may yield a new generation of superfast computer chips and ultrasensitive molecular detectors

The Promise of PLASMONICS

By Harry A. Atwater

LIGHT BEAM striking a metal surface can generate plasmons, electron density waves that can carry huge amounts of data. If focused on a surface etched with a circular groove, as in this artist's rendering, the beam produces concentric waves, organizing the electrons into high- and low-density rings.

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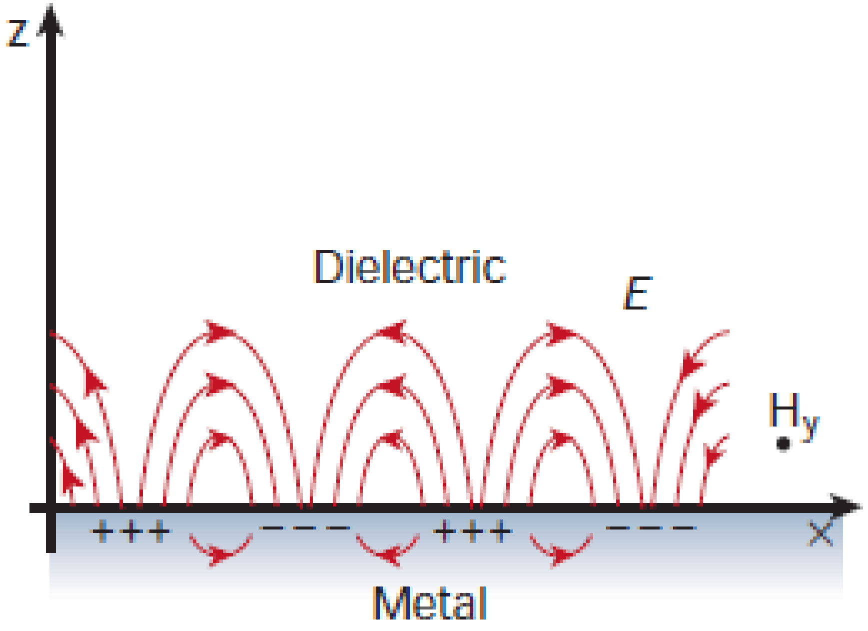
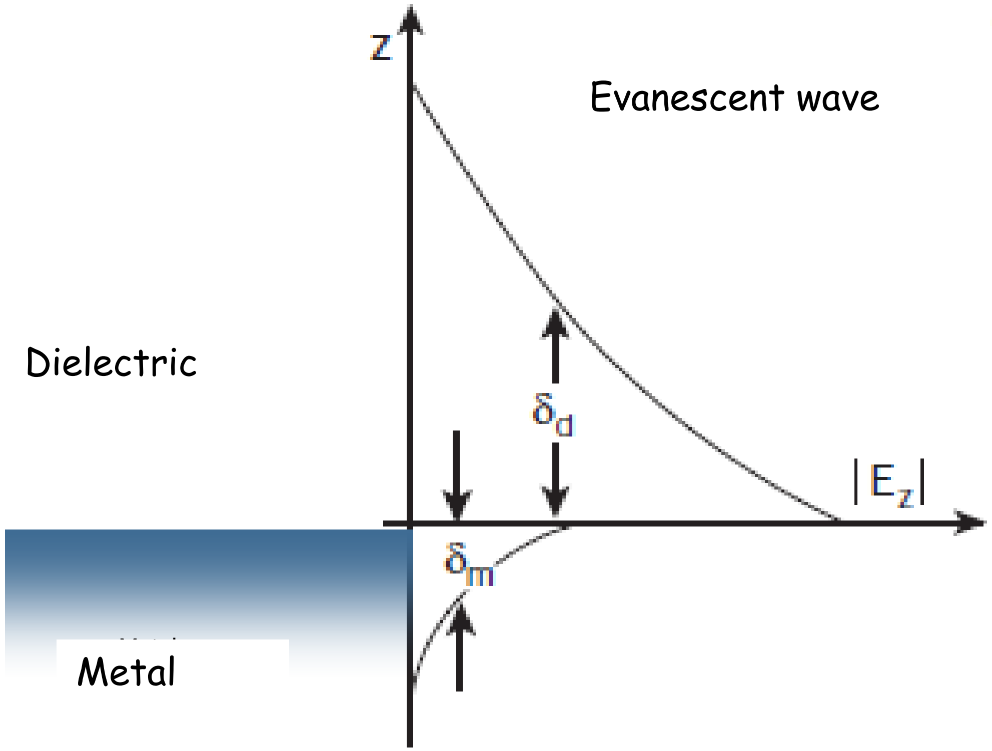
What are surface plasmons?



$\mathrm{Re}[\varepsilon_m]<0$

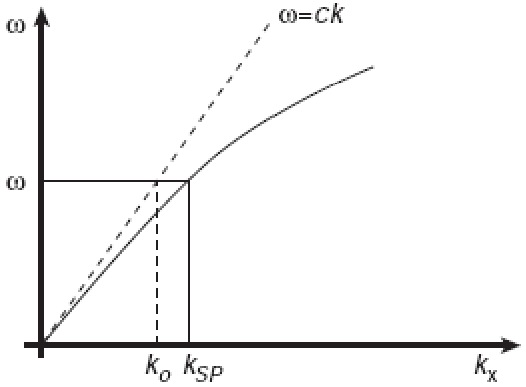
$\mathrm{Re}[\varepsilon_d] < -\mathrm{Re}[\varepsilon_m]$



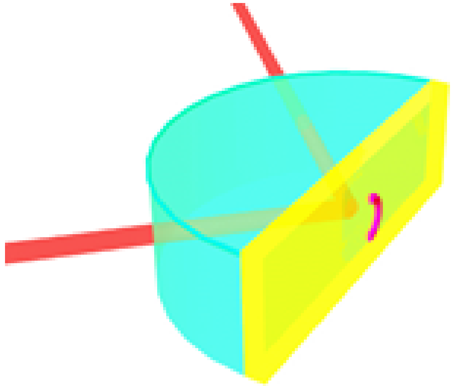
How can they be excited?

Due to their bounded nature, the momentum of surface plasmons is always larger than the moment of light. Thus in order to excite surface plasmons using light it is necessary to use coupling strategies:

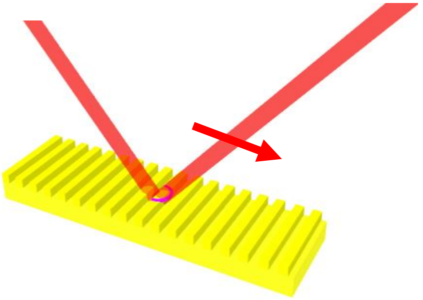
$$k_{SP} = k_0 \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}} > k_0$$



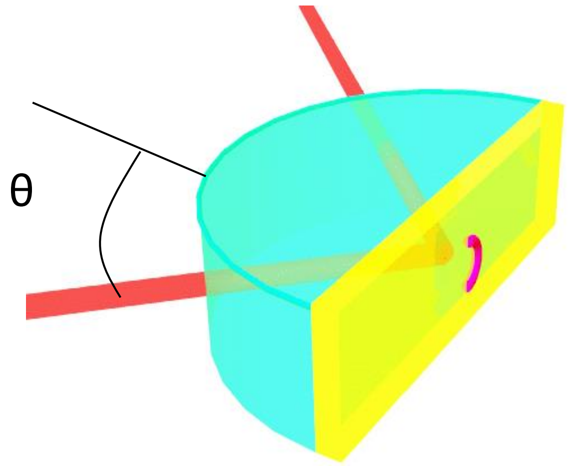
Attenuated total reflection
(Kretschmann configuration)



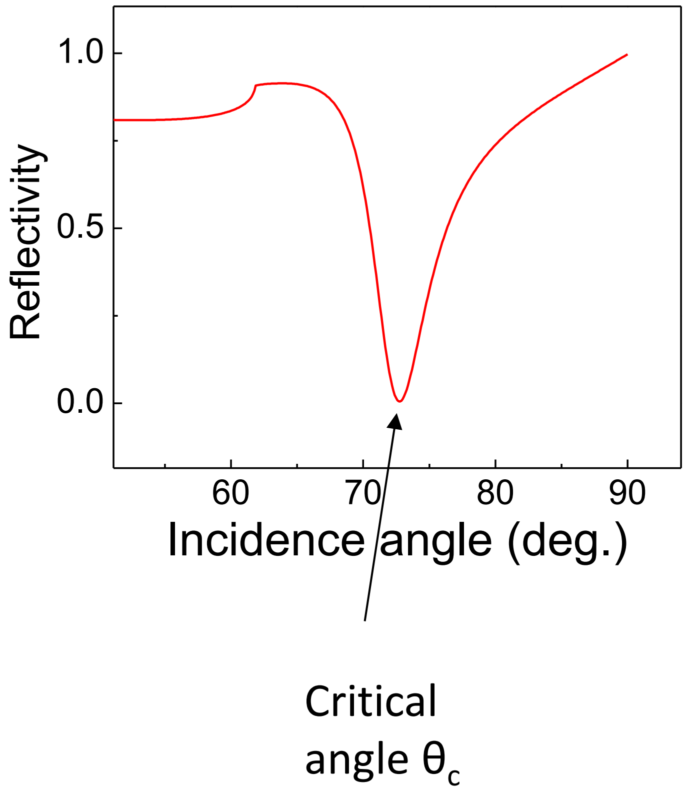
Gratings



What is the effect of the excitation of surface plasmons?

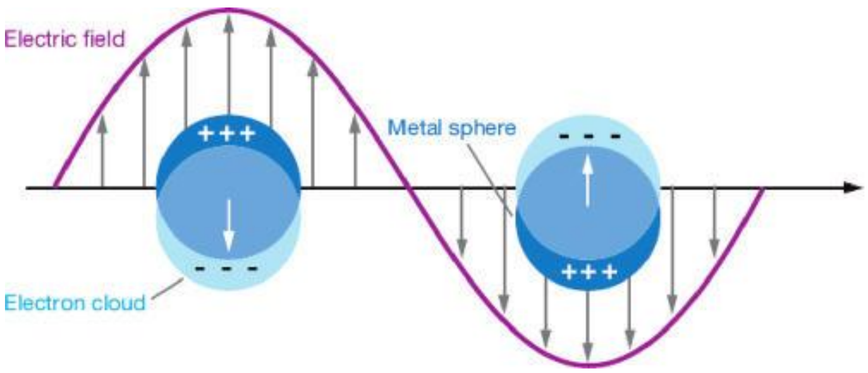


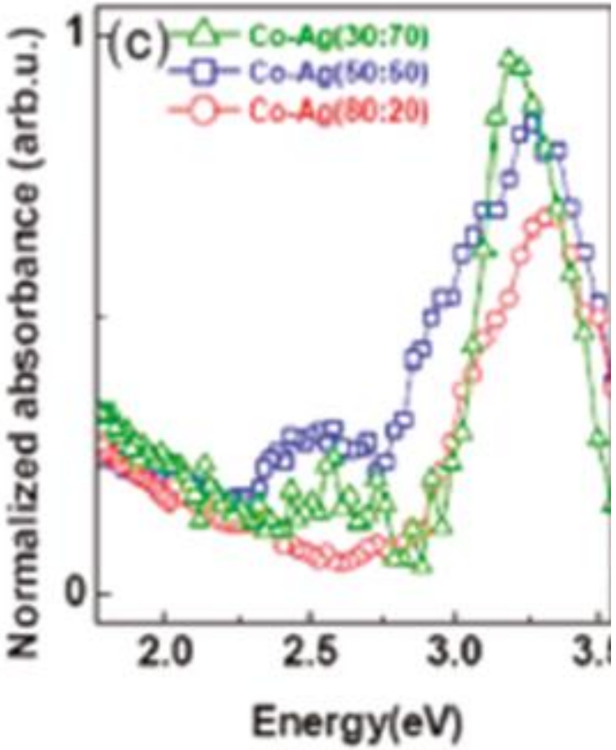
$$k_{SP}=k_0\sqrt{\frac{\varepsilon_d\varepsilon_m}{\varepsilon_d+\varepsilon_m}}=k$$

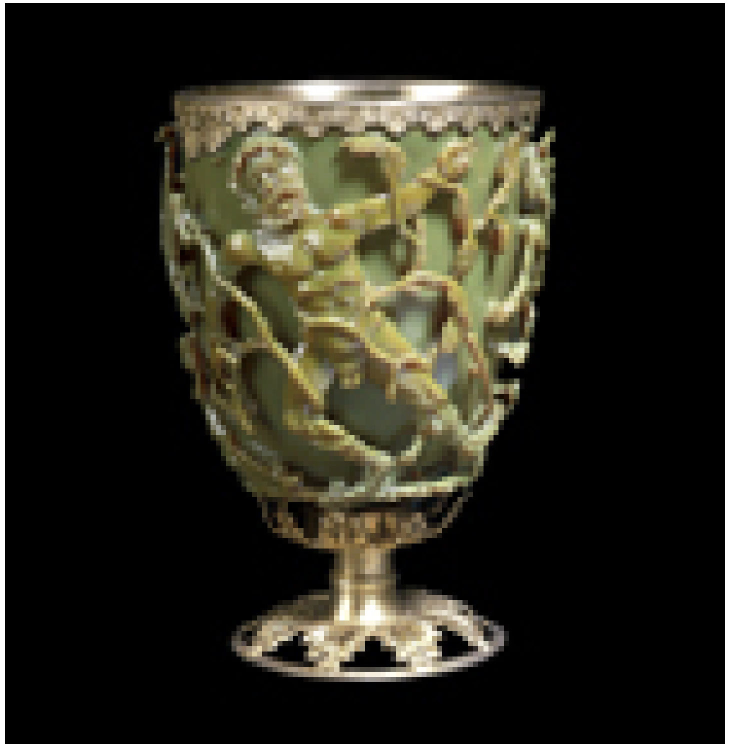


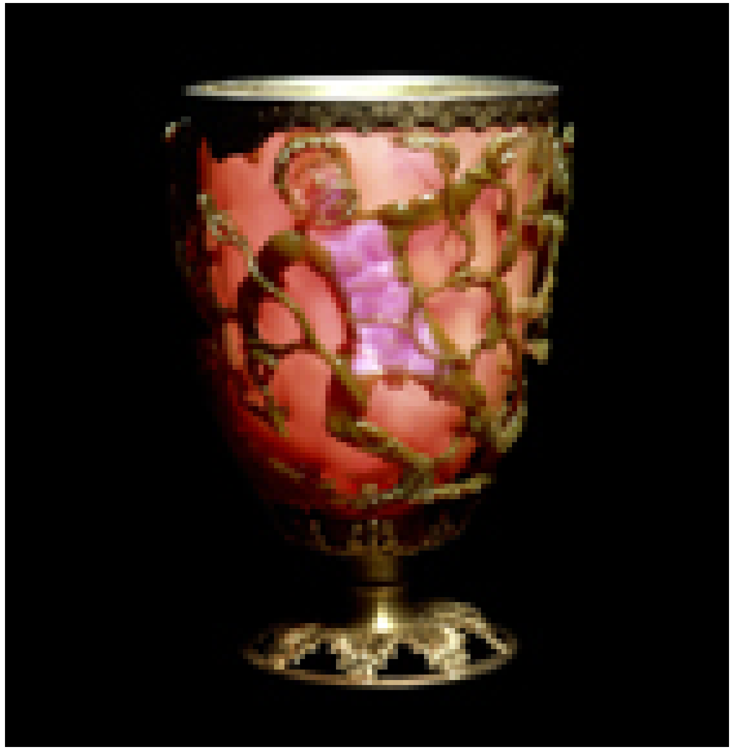
Localized Surface Plasmons

Excitation of LSP by an electric field (light) at an incident wavelength where resonance occurs results in strong light absorption, and an enhancement of the local electromagnetic fields. Associated with the resonance it is observed a strong increase of the absorbance (decrease of the transmittance) at specific frequencies.

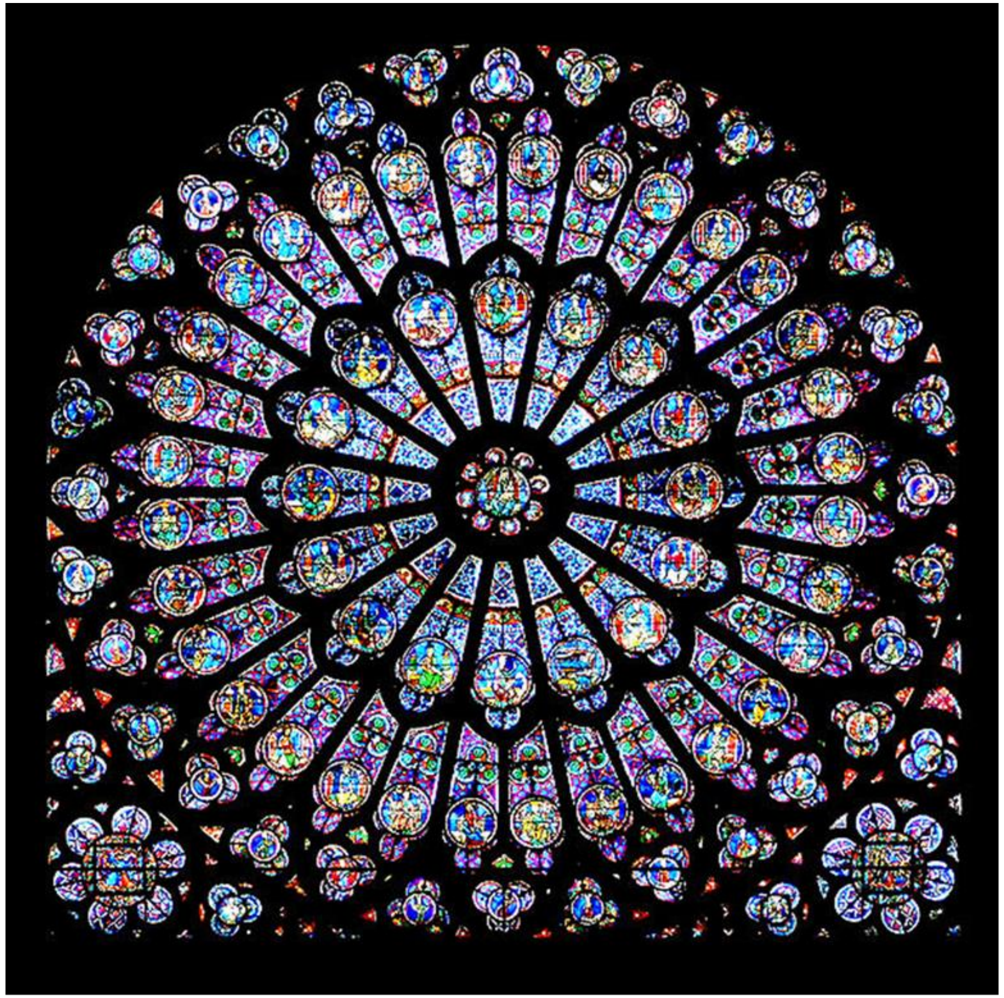








4th Century: The Lycurgus Cup (Rome) is an example of dichroic glass; colloidal gold and silver in the glass allow it to look opaque green when lit from outside but translucent red when light shines through the inside



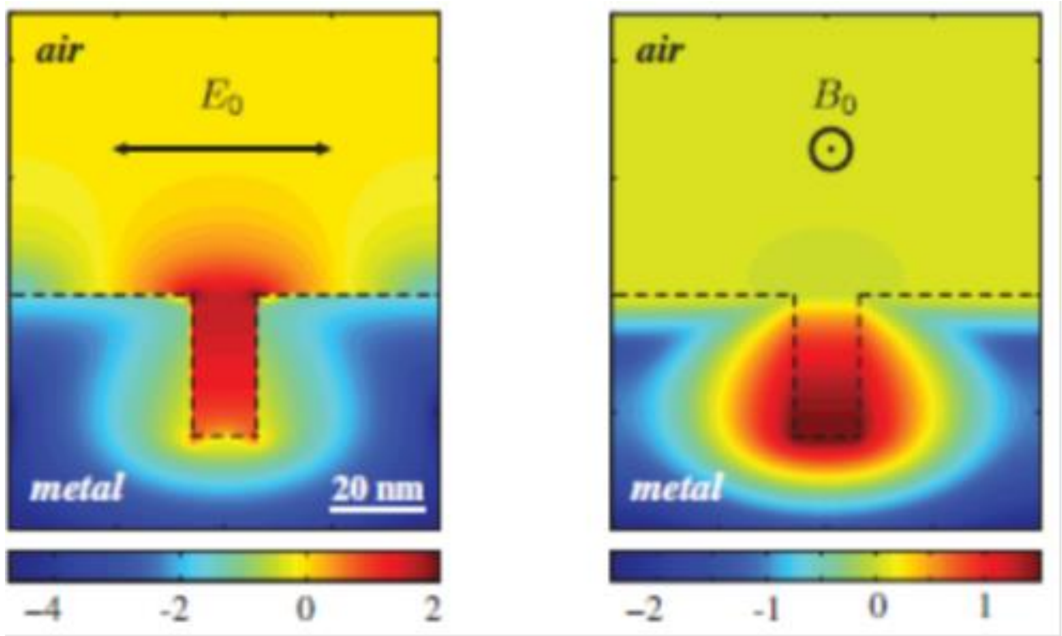
Gothic stained glass rose window of Notre-Dame de Paris.
The colors were achieved by colloids of gold nano-particles

Surface plasmon resonance approach

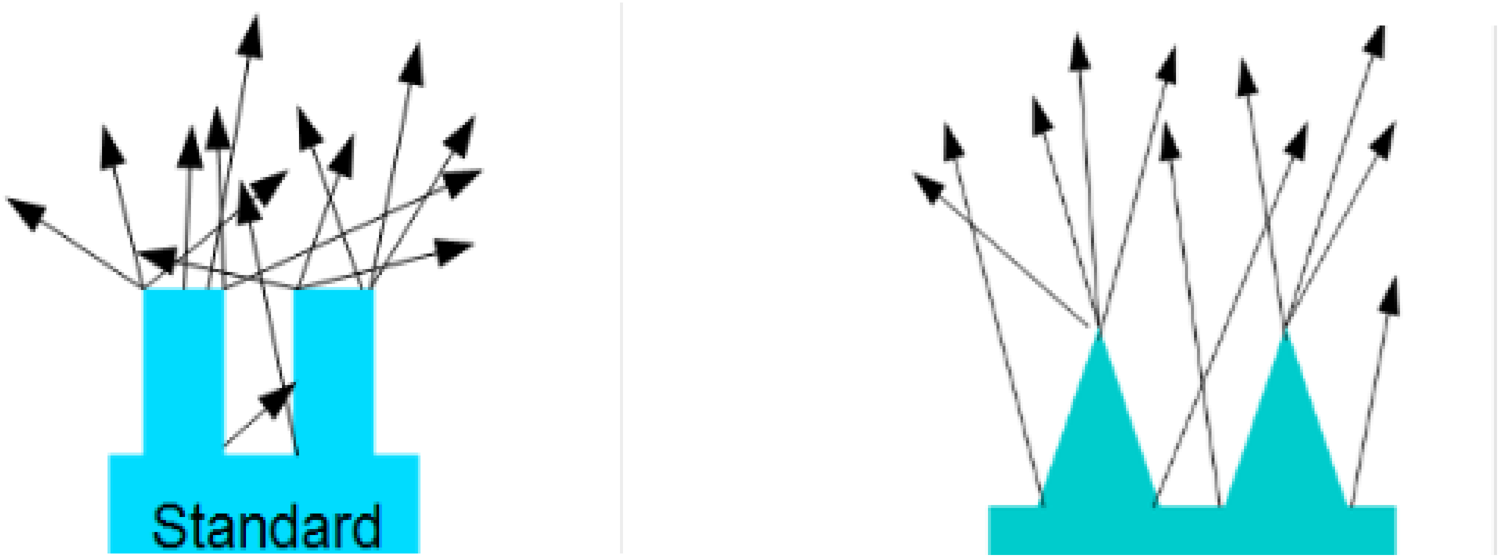
- The only way to increase the QE in a metallic photocathode without adversely affecting ε is to decrease the optical penetration depth and reduce the metal reflectivity. Coupling to the SPPs modes on the metal surface is the ideal solution for this problem.
- It is possible to design a system where light couples to the SPP modes on the surface yielding a perfect absorber at a chosen wavelength. Another advantage for photoemission is that an SPP has shorter evanescent fields than optical skin depth.
- Strong QE increase was demonstrated using this approach by Callcott et al. [Callcott, T. A. "Volume and surface photoemission processes from plasmon resonance fields." Phys. Rev. B (1975)]

Present state of research

- Metal-based Plasmon enhanced photocathodes yielding somewhat higher quantum yield compared to flat metallic surfaces have been demonstrated. [R. K. Li, H. To, G. Andonian, J. Feng, A. Polyakov, C. M. Scoby, K. Thompson, W. Wan, H. A. Padmore, and P. Musumeci, "Surface-Plasmon Resonance-Enhanced Multiphoton Emission of High-Brightness Electron Beams from a Nanostructured Copper Cathode", Phys. Rev. Lett. 110, 074801 (2013)].
- Such nano-engineering photoemitters can have significant advantages of lifetime and robustness over semiconductor photocathodes, but it is worth noting that typical designs have shown large spreads of energy and emittance which has a deleterious effect on the overall beam brightness.



Our approach



- *(Left)* typical pattern explored so far. *(Right)* improved tailored profile by using conformal thin film deposition onto gratings.

Surface Plasmon Resonance on gratings

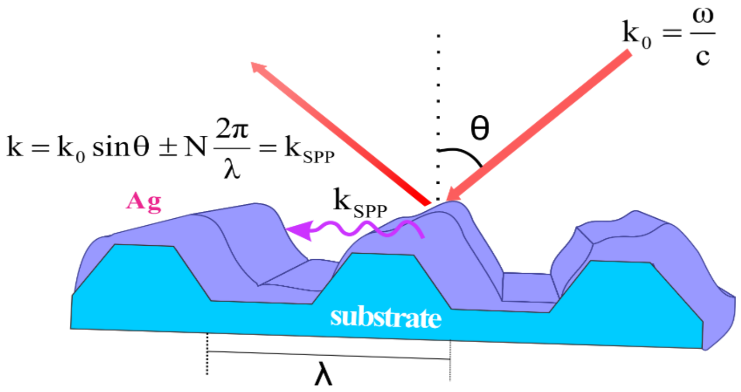
- Electrons resonate at dielectric-metal boundary.

  Dielectric constant requirement:

  $\varepsilon_1 < 0$ and $|\varepsilon_1| > \varepsilon_2$

- Excitation of resonance need to enhance the momentum of incident light.
- SPR enhanced photoemission has been detected 9 times of that of planar Al photocathodes under deep UV light.[1] Higher enhancements (~E3 times) of charge yield have been reported under multi-photon process.[2]

$$k_{spp} = \frac{w}{c}\left(\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right)$$



[1] Y. Watanabe, W. Inami, and Y. Kawata, *J. Appl.Phys.* **109,** 023112 **(**2011**)**
[2] R. K. Li, *et al.*, *P.R.L.* **110**, 074801 (2013)

Grating coupler excited surface plasmon resonance

- Thus, on a continuous metal thin film, the electrons will oscillate coherently when the incident light wave vector is enhanced to match the dispersion relationship (in air/vacuum):

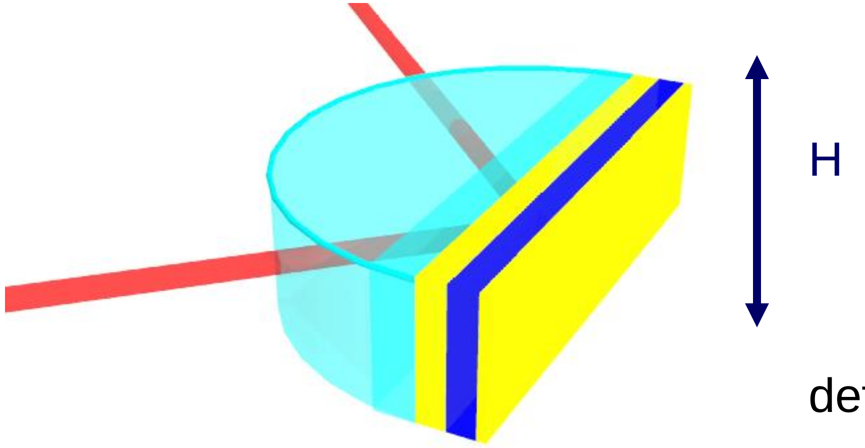
$$k_{spp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1}{\varepsilon_1 + 1}}$$

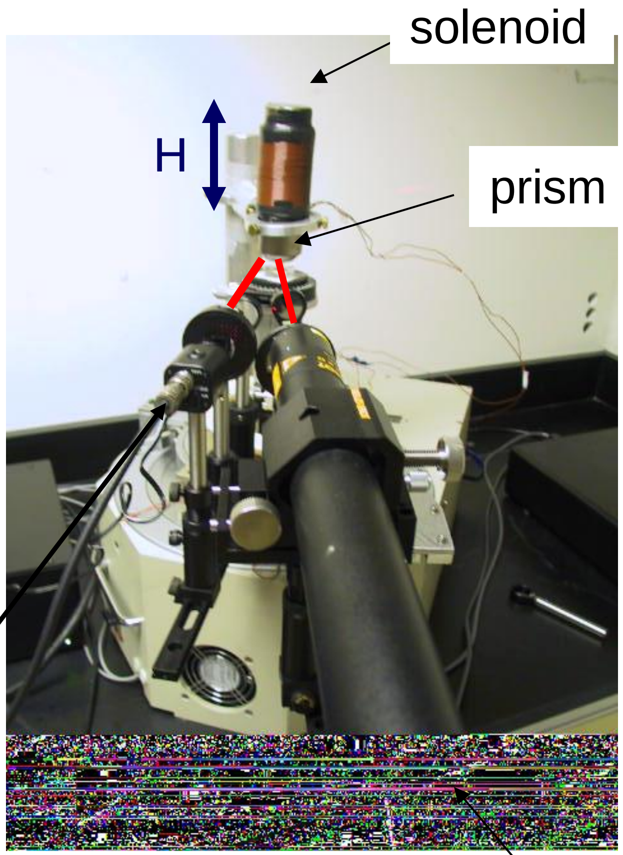
- To match this dispersion requirement, one can use grating coupler to enhance the wave vector of incident light.

$$k_{spp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1}{\varepsilon_1 + 1}} = \frac{\omega}{c}\sin\theta \pm N\frac{2\pi}{\lambda_g}$$

Characterization of-plasmonic materials

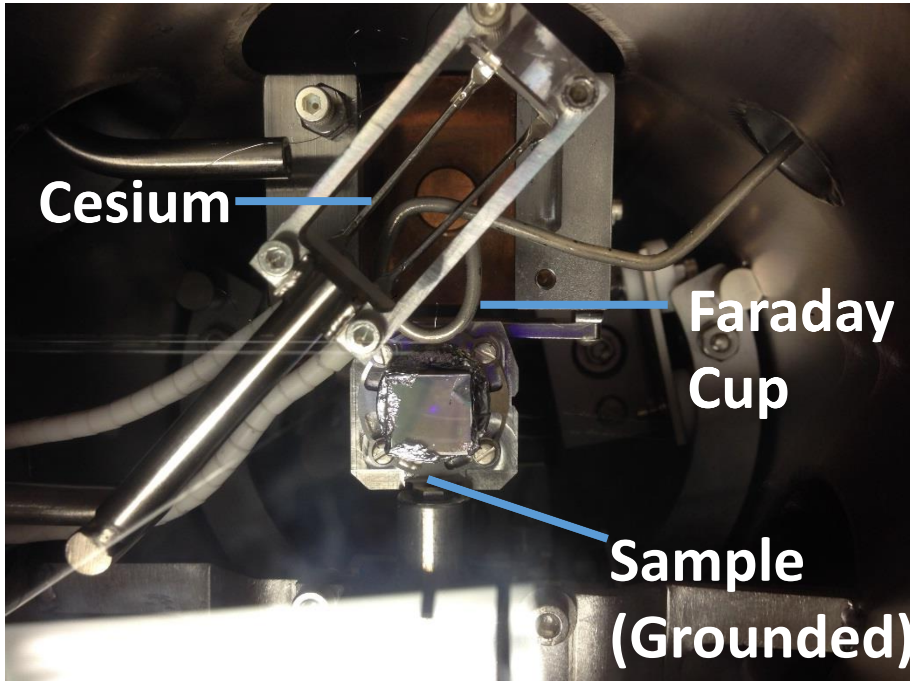
- Ellipsometry
- X-ray diffraction
- MO-SPR set up

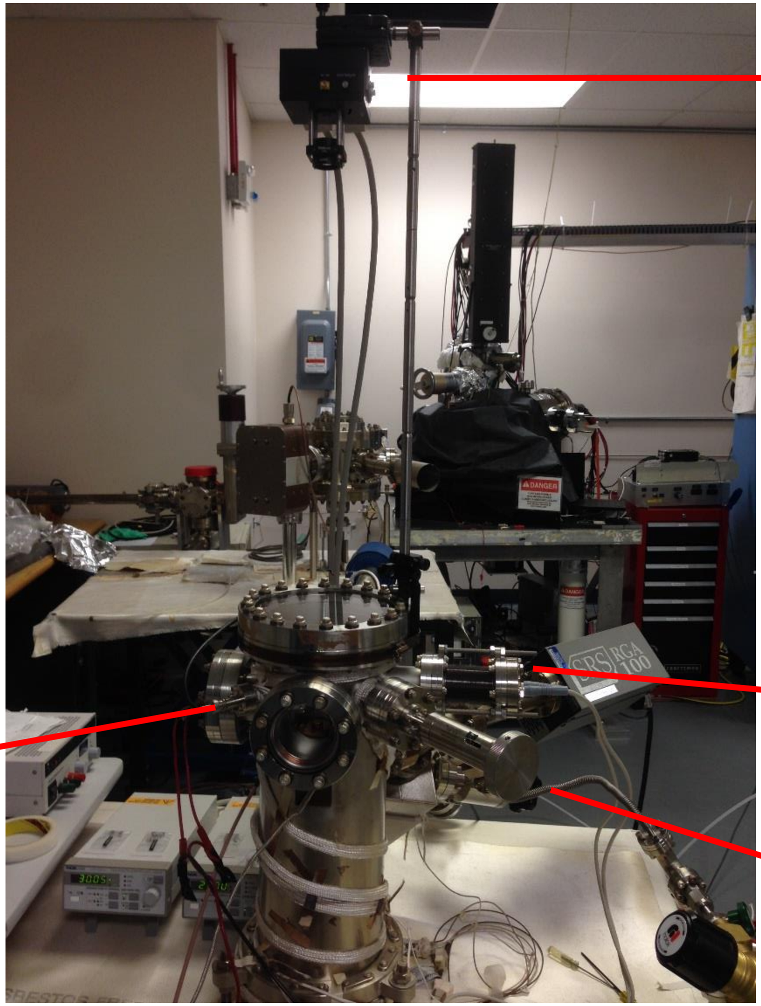




Resolution 10^{-3} deg.

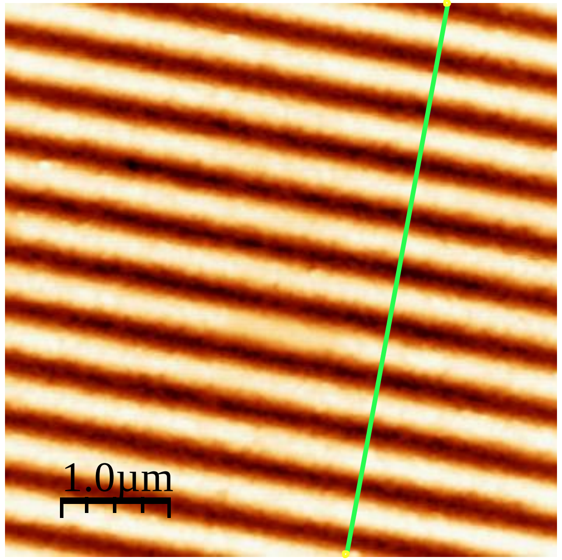
Photo-current Experimental Setup



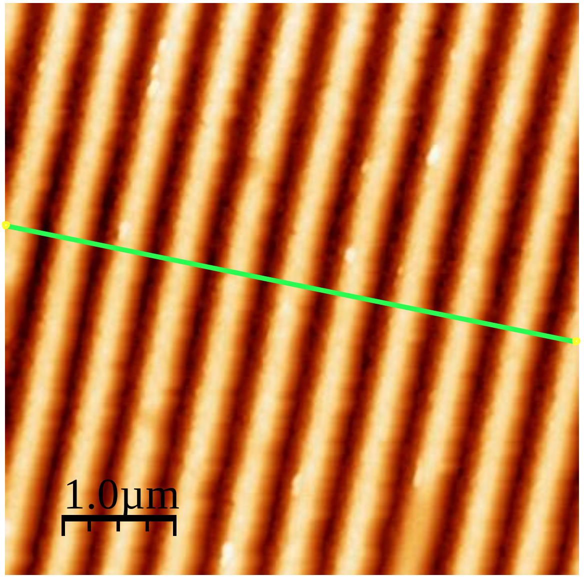


Topography and line-scan profile on bare grating

Sample @ 0 deg



Sample @ 90 degs

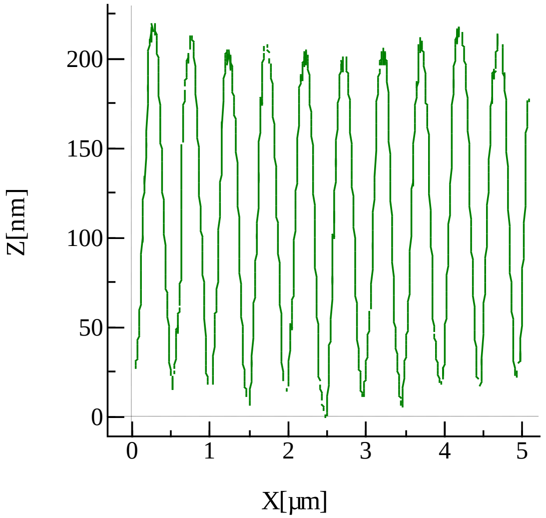


Avg periodicity: 488.2 nm

Error from standard ~ 12.1%

Avg groove depth: 209.3 nm

Error from standard ~ 3.98%

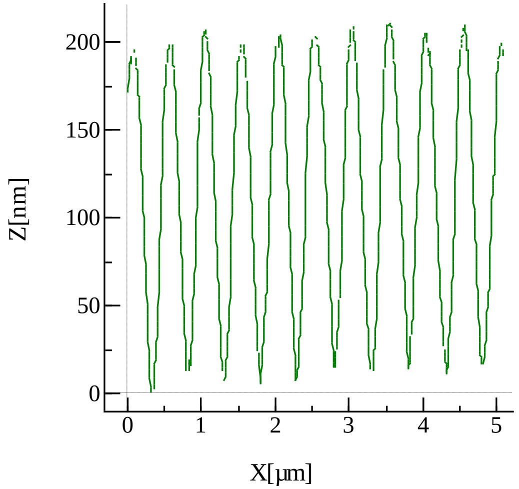


Avg periodicity: 500.1 nm

Error from standard ~ 9.97%

Avg groove depth: 206.4 nm

Error from standard ~ 2.48%



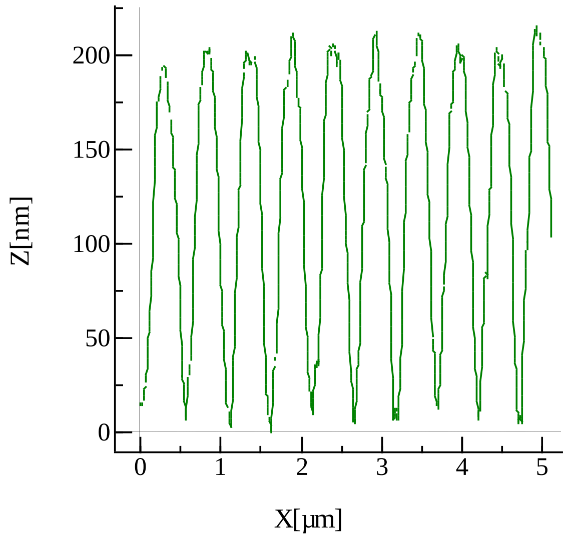
Topography after Ag deposition of 50nm

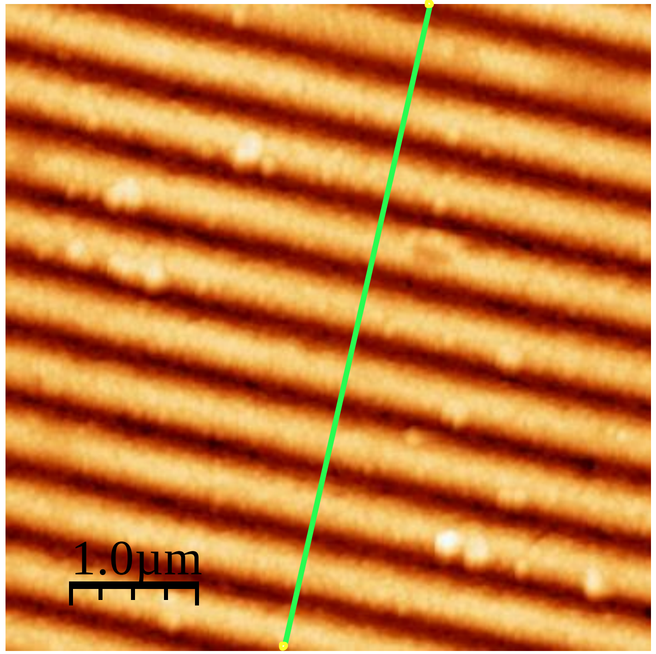
Avg periodicity: 525.0 nm

Error from standard ~ 5.49%

Avg groove depth: 205.8 nm

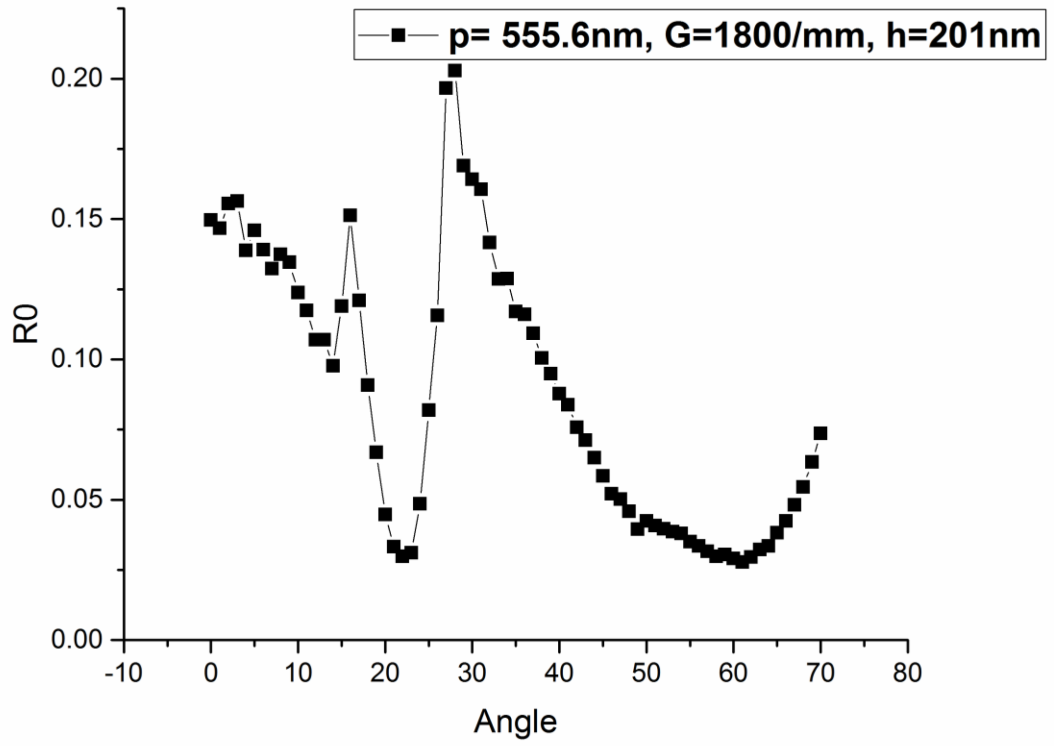
Error from standard ~ 2.38%



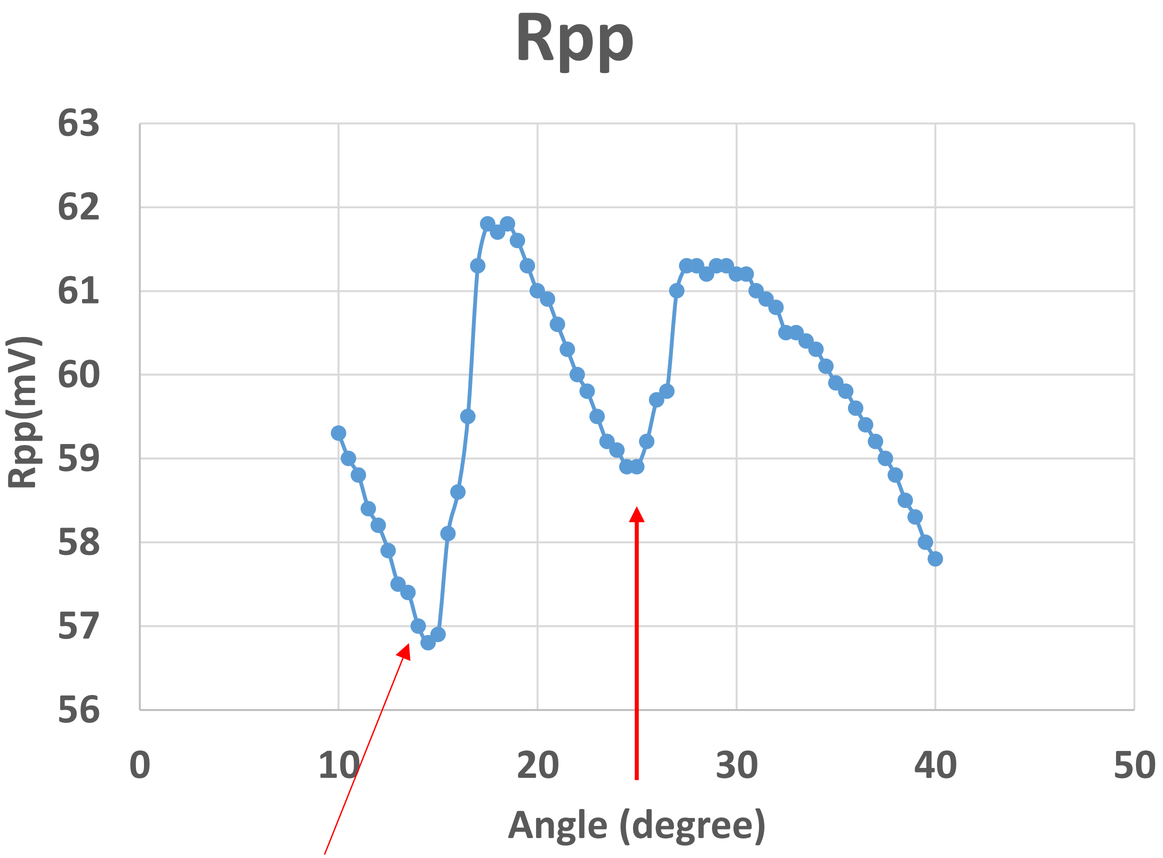


SPR measurement on glass grating sample

Simulation Result



SPR @ ~22 degs



Also shown in the simulation, but the reflectivity reformed.

SPR @24.5 deg;

After Cs deposition: Cs/Ag50nm/Al/Glass (ND)

<u>Grating Standard Information:</u>

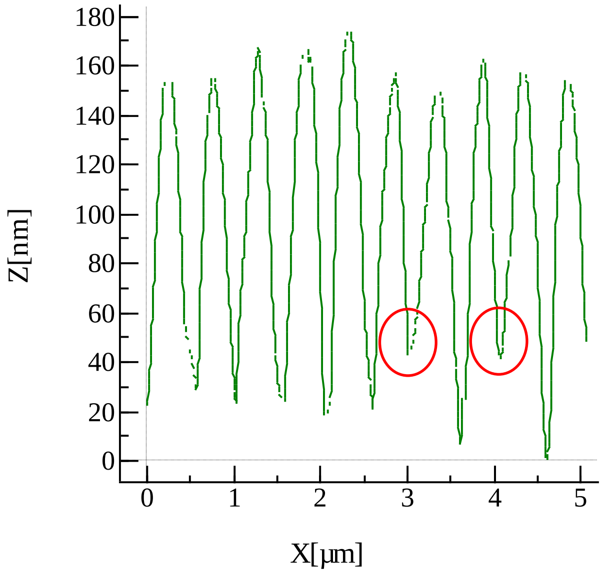
Groove depth: 209nm

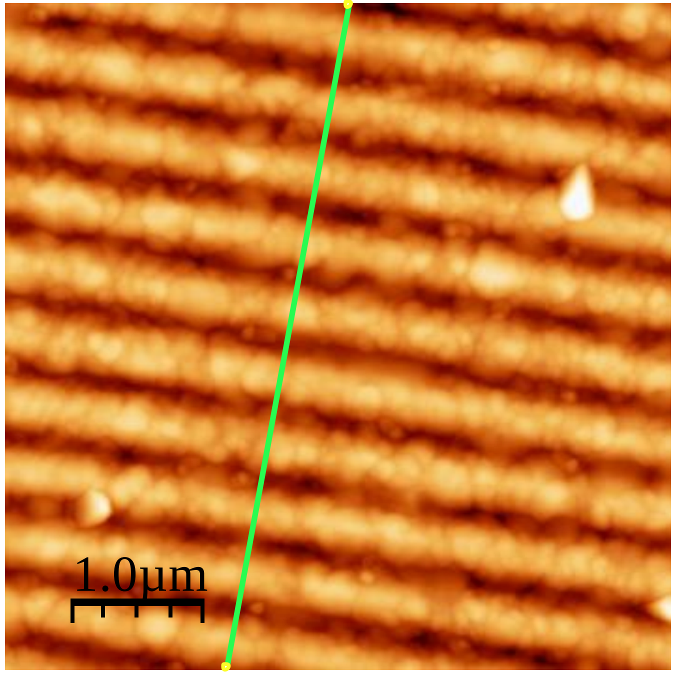
Periodicity: 555.5nm

<u>AFM measurement:</u>

Avg groove depth: 161.76 nm

Avg periodicty: 504nm

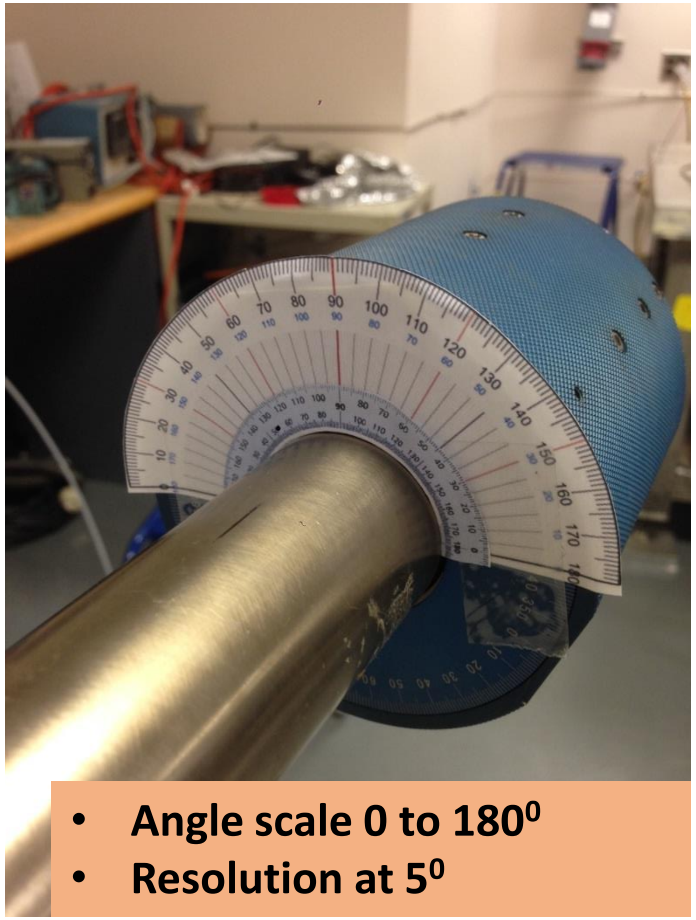




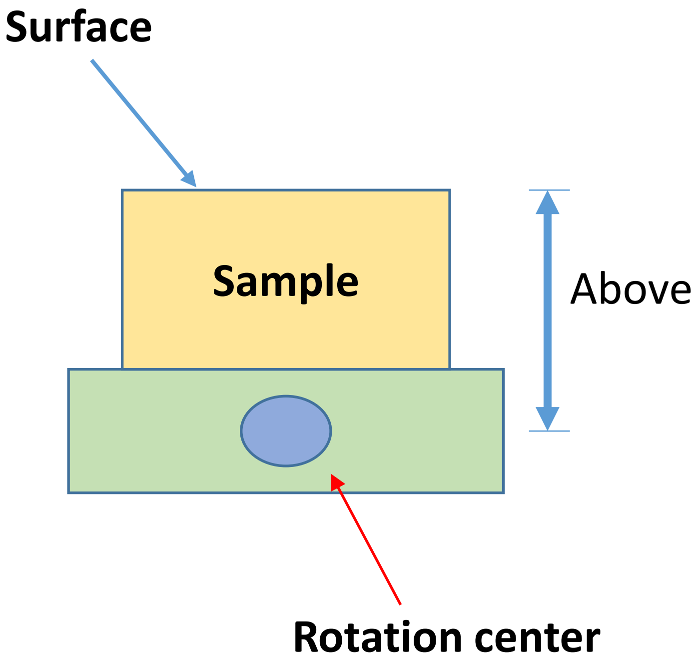
Groove depth has been decreased because of the Cs layer. Groove depth also shows a larger variation than w/o Cs.
<u>QE ~ 0.1~0.178%.</u>

Laser setup

Laser alignment



- Angle scale 0 to 180^0
- Resolution at 5^0



Angle dependence of cesium activation

	Position I	Position II	Position III	Position IV
Maximum current (nA)	75 @110^0	85 @55^0	73 @117^0	86 @51^0
Minimum current (nA)	57 @67^0	57 @107^0	57 @65^0	55 @99^0
Difference in current (%)	31	51	28	56

Lifetime improvement

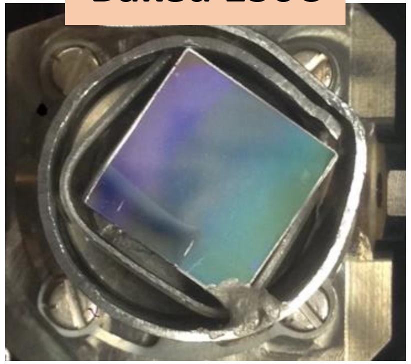
- **Sample description**: Ag55nm/Al/Glass grating (without annealing); initially baked with loadlock under **150C** to reach high vacuum and showed less oxidation on cathode surface. <u>This gives a cleaner surface than that previously baked under 200C.</u>

- **Previous results**: Only can activate the cathode by Cs. QE ~0.26% maximum at "direct" incident angle. Lifetime ~ 10 to 20 hrs.

- **Current update**: QE~0.25% with Cs. Lifetime improves to 49hr. However, after heat cleaning the surface, the QE with Cs dropped somewhat to 0.14~0.15%.

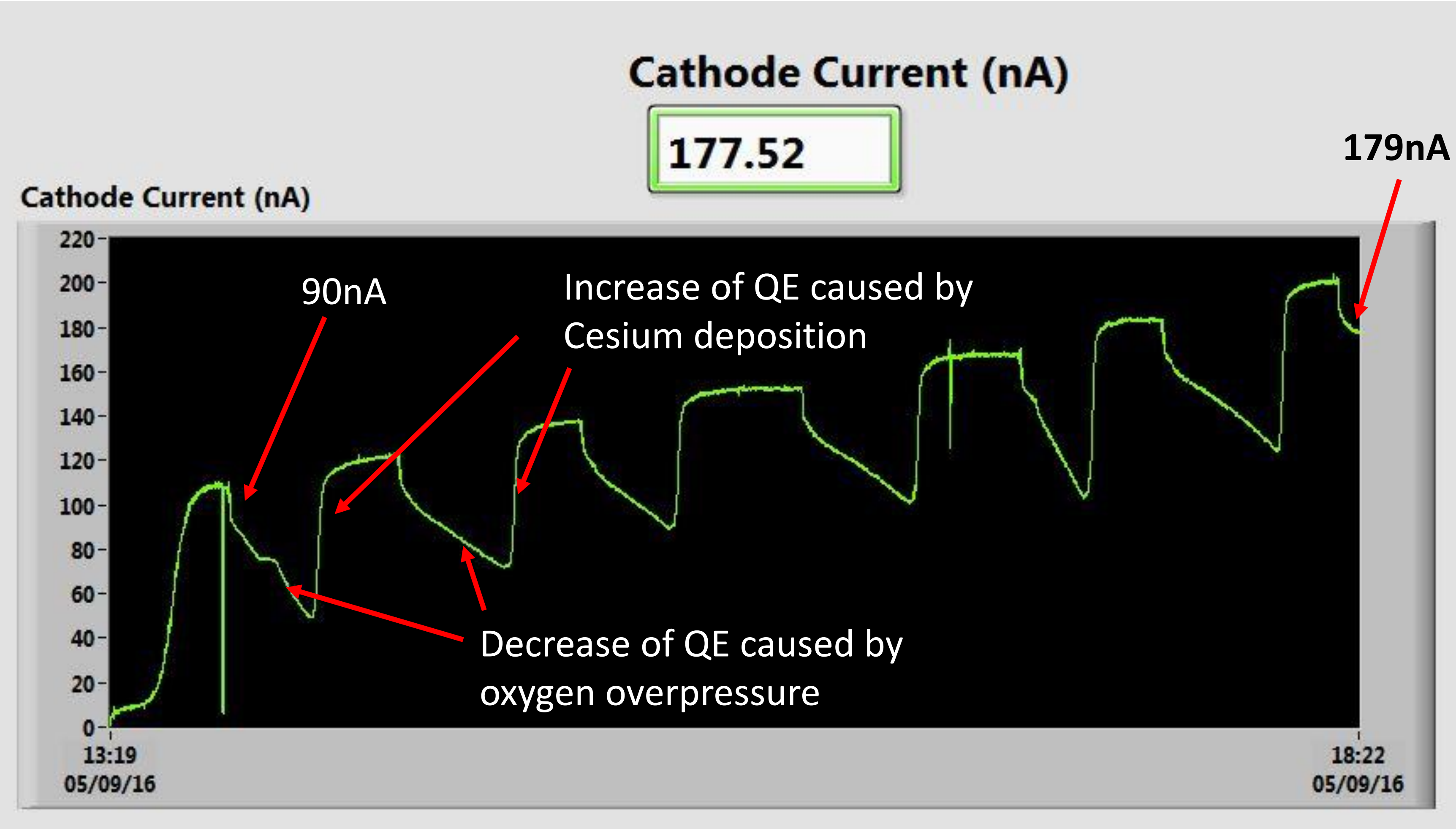
- Found suitable oxygen pressure to form Cesium oxide(s) on this sample.

Baked 150C



Baked 200C

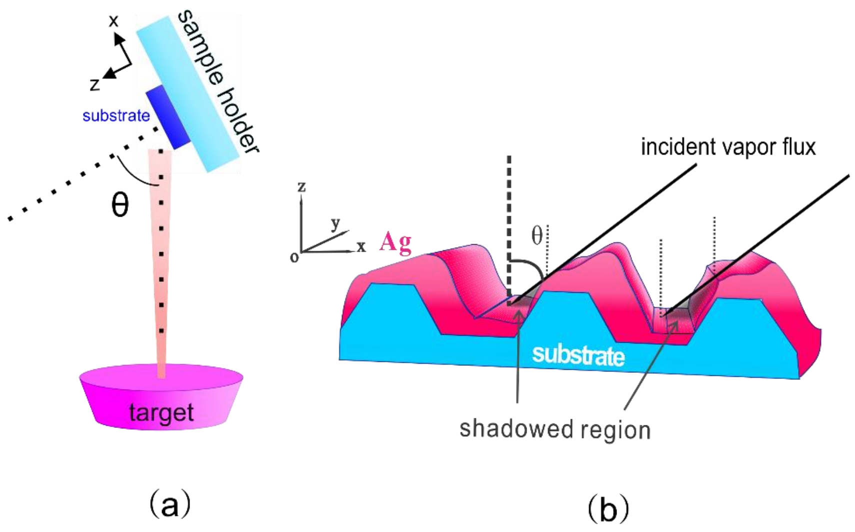
Oxygen Overpressure yo-yo curve activation



- **QE after first cesium deposition ~ 0.15% (heat-baking degradation)**
- **After 6 cycles QE ~ 0.28%. (1.87 times)**
- **Life time is ~48hrs.**

Present status of our efforts

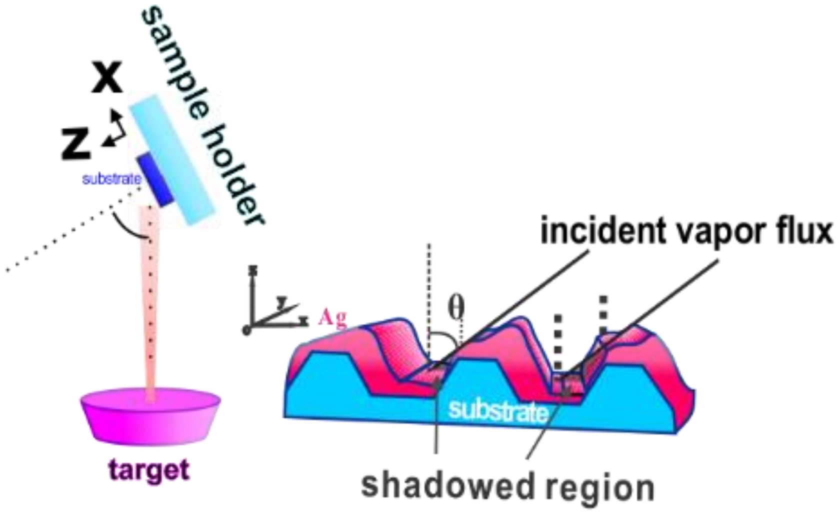
- **<u>The angle dependence</u>** of a grating silver cathode shows a maximum of 50% enhancement of QE.
- **<u>Silver surface contamination</u>** can affect the cathode lifetime. By baking under suitable temperature, the cathode shows a steady lifetime of ~48hrs.
- **<u>Substrate annealing</u>** indicates improvement on the QE performance of the cathode from 0.26% to 0.4% for cesium activation.
- **The plan is to attempt oblique incidence deposition (OAD) which is expected to further enhance local EM fields and hence QE.**

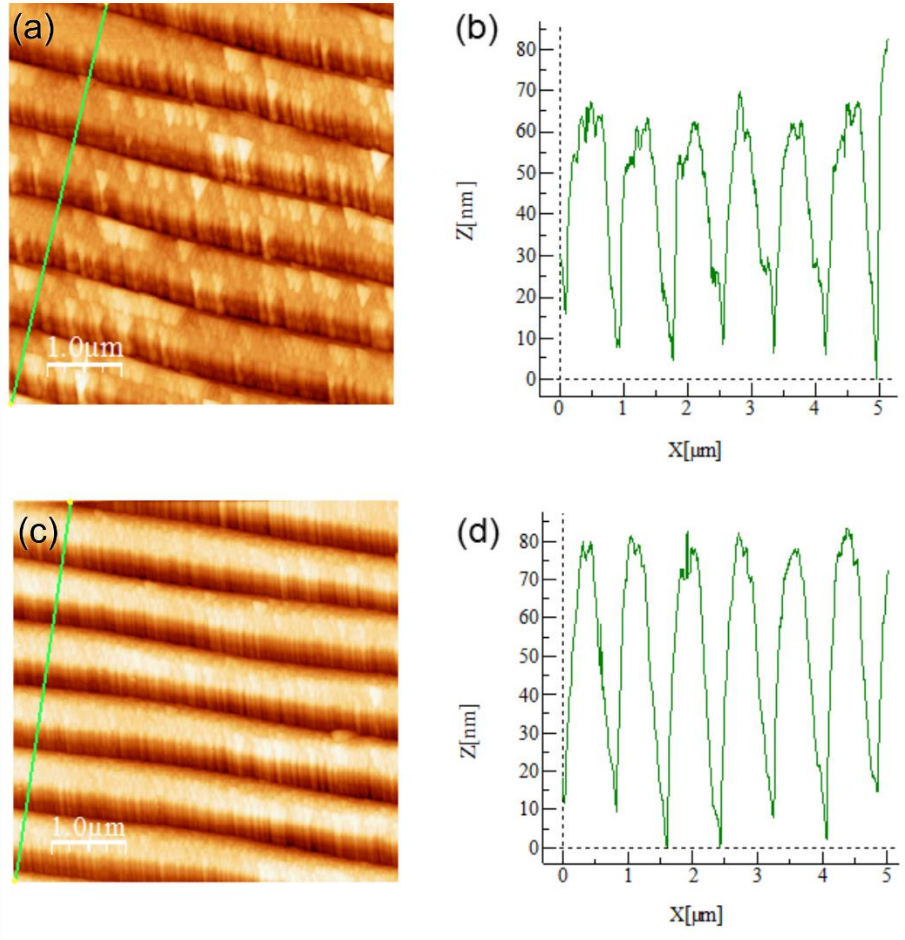


By applying OAD it is possible to alter the grating structure of the cathode and enhance the SPR induced EM field.

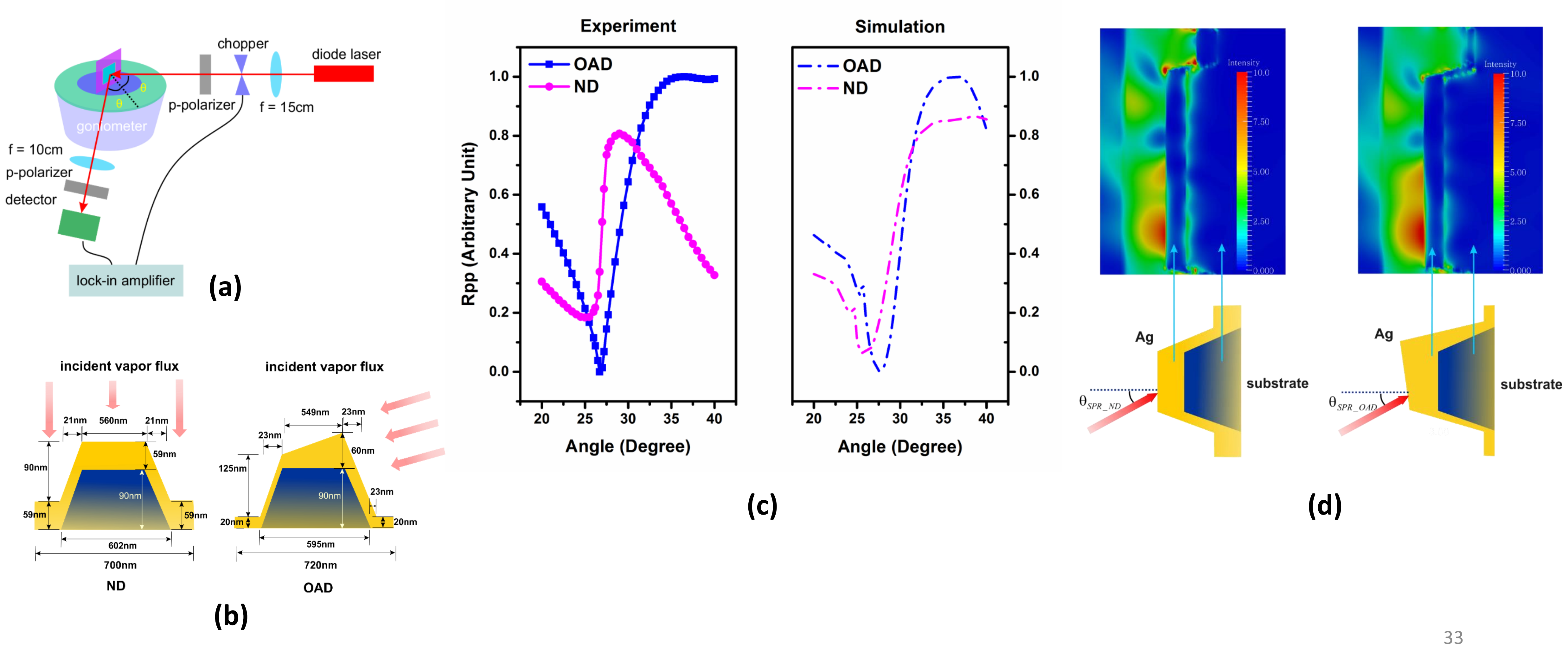
OAD Method

- Large Incident Angle: ~77 degrees versus normal incident angle. Ar pressure at 7.5 x 10^{-3} Torr. Base pressure 2.0 x 10^{-6} Torr
- AFM topography shows good coverage of silver thin film.





SPR Enhancement by OAD

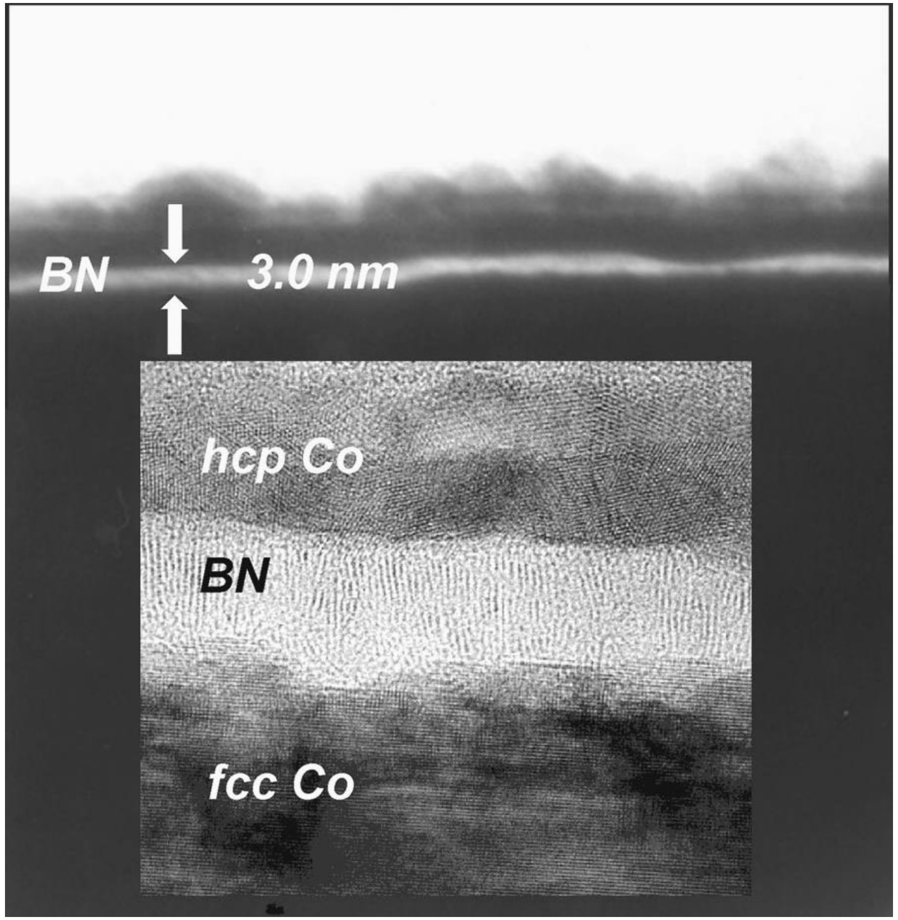


On spin-polarization

- Potential alternatives to GaAs-based polarized photocathodes are the half metallic ferromagnets (HFM). These are materials with an unusual band structure. At the Fermi energy they have a band gap for one spin band and are metallic for the other, characterizing them by a theoretical 100% spin polarization in the ideal case where the temperature is T= 0 K and spin-orbital interactions are neglected.
- Recently, the validity of band structure calculations of some Heusler compounds has been experimentally tested by Ultraviolet Photoemission Spectroscopy (UPS). First measurements on Co2MnGa thin films under these conditions resulted on a spin polarization of 60% at the Fermi energy, which is the highest experimental spin polarization value obtained for this Heusler compound. This value is quite close to the theoretical prediction of 67 %. Many other Heusler compounds have also been investigated using spin polarized tunneling magneto –resistance ratio (TMR) to characterize the actual spin polarization achieved

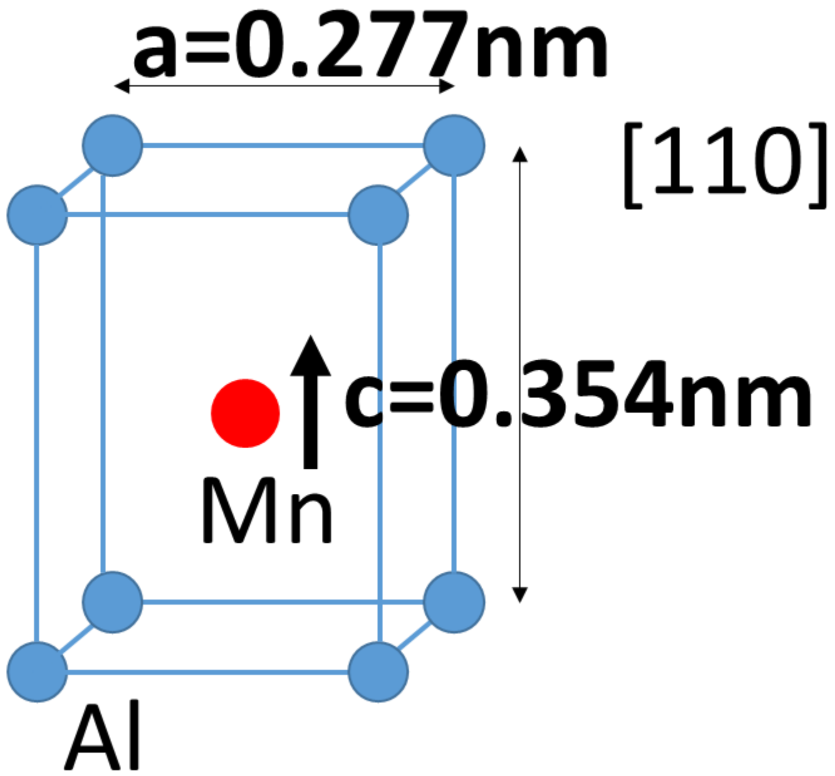
Spin-polarized structures

- We have also worked on magnetic thin films and multilayers in particular for spin-dependent tunneling applications aiming at high TMR.
- For these studies we explored magnetic materials with large perpendicular magnetic anisotropy and insulating tunneling layers, including MgO, since the latter has been shown to lead to the highest TMR.
- This approach suggests the possibility of highly polarized electron injection into the vacuum through a suitable insulating overlayer.



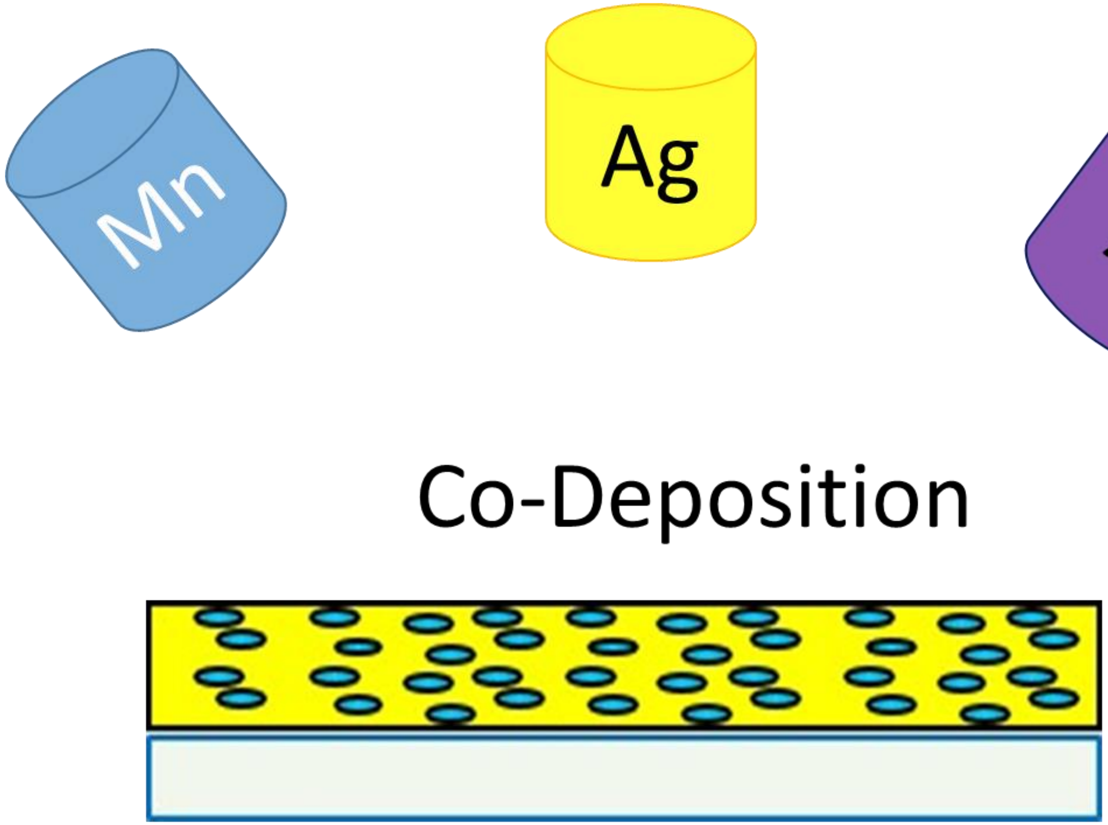
Silmanal

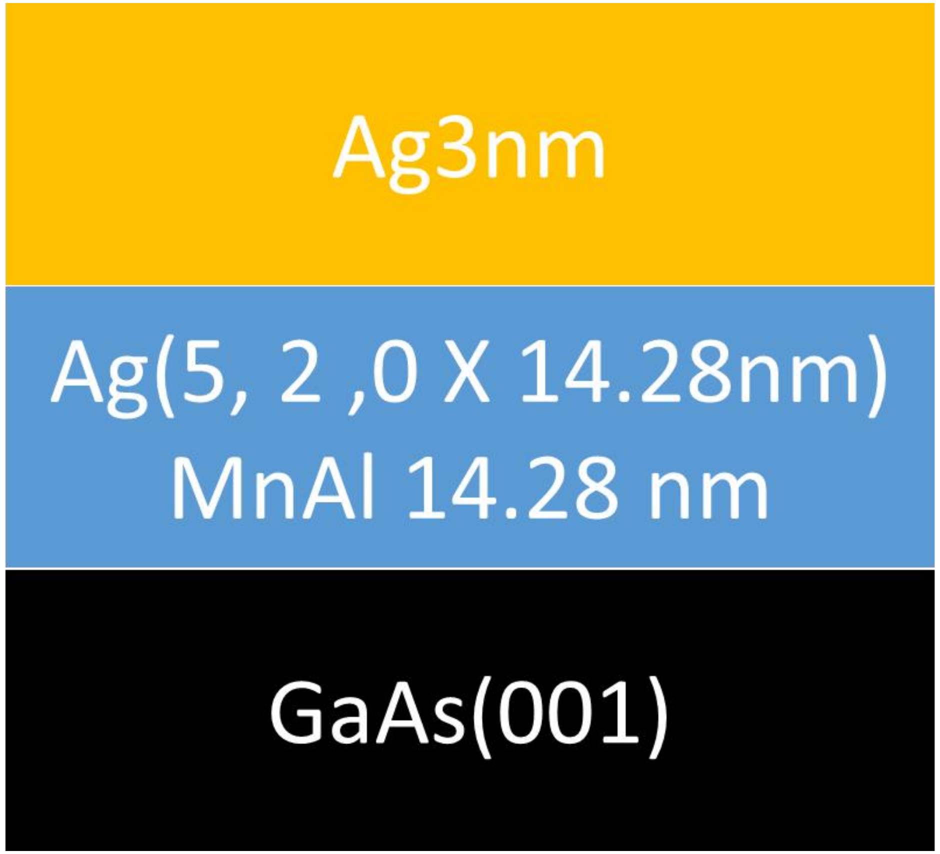
- Silmanal (Ag2MnAl) is a ternary alloy of Ag, Mn and Al and it was the first known magnet of record that has exceptional coercive force, around 6300 Oe , due to chemically ordered MnAl clusters embedded in the silver matrix. The Silmanal alloy consist of 86.6% Ag, 8.8%Mn and 4.4%Al.
- We have demonstrated the magnetic properties of ordered MnAl films and continue working on correlated microstructure-magnetic properties studies of AgMnAl films of various compositions to optimize the films for the present application.
- The Ag matrix is expected to aid in the SPR excitation while the τ phase in the MnAl clusters is expected to provide the spin-polarization.



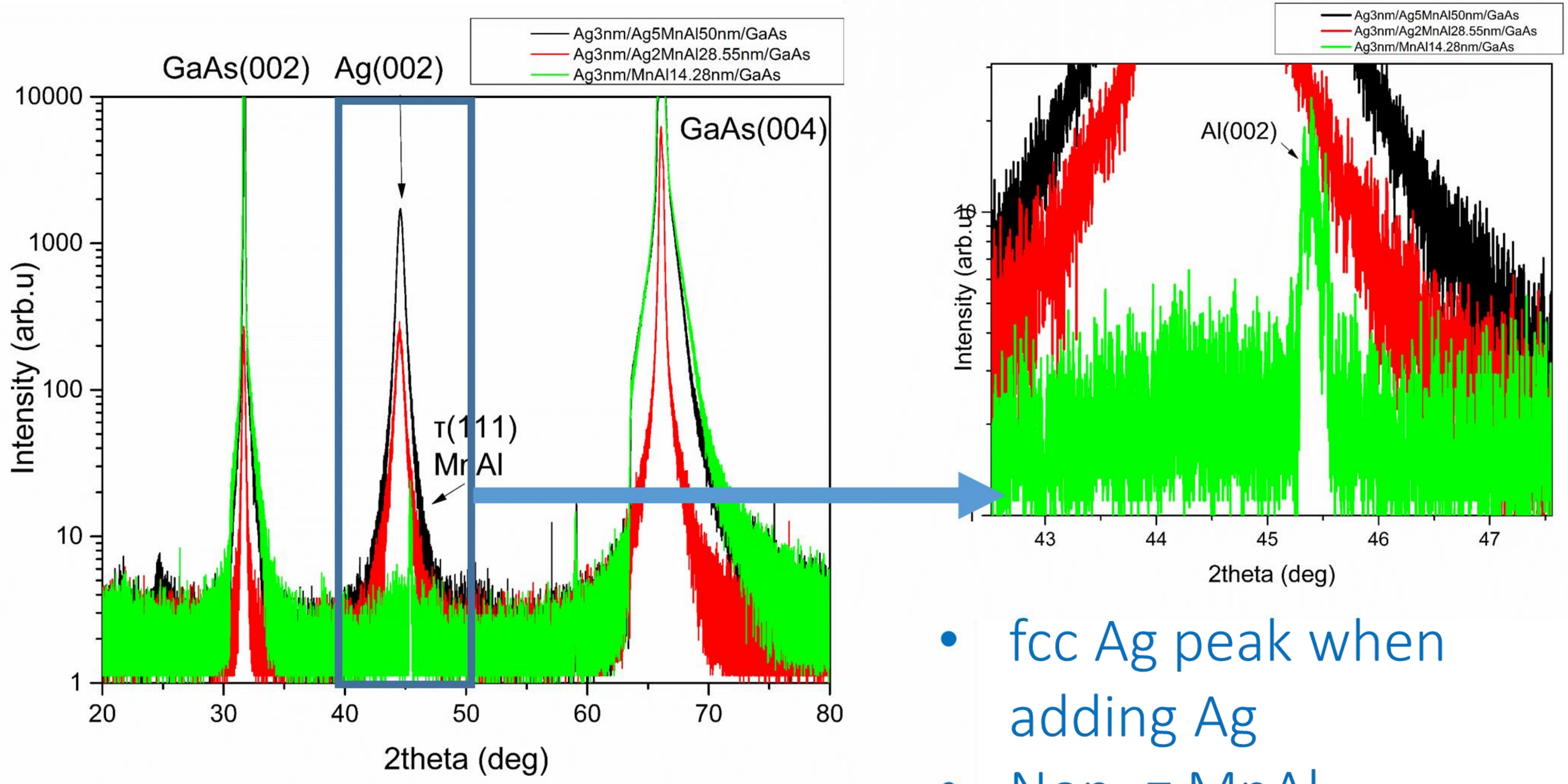
τ phase

Silmanal thin film deposition





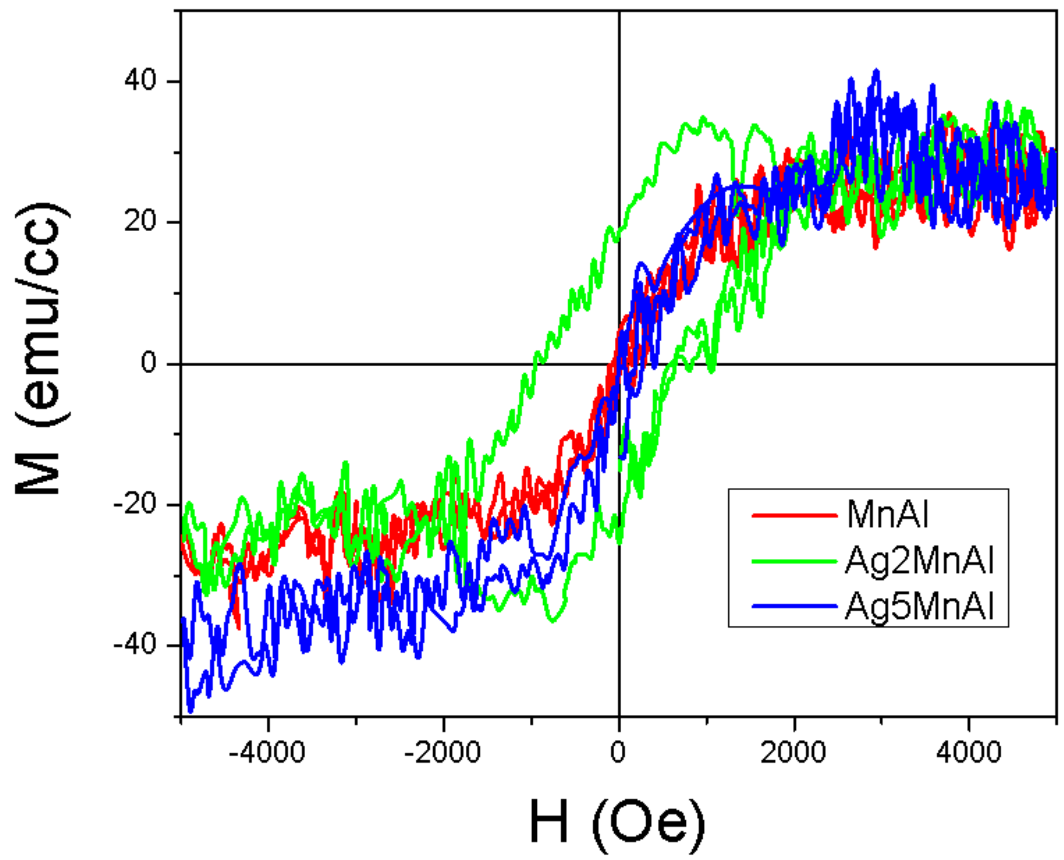
XRD



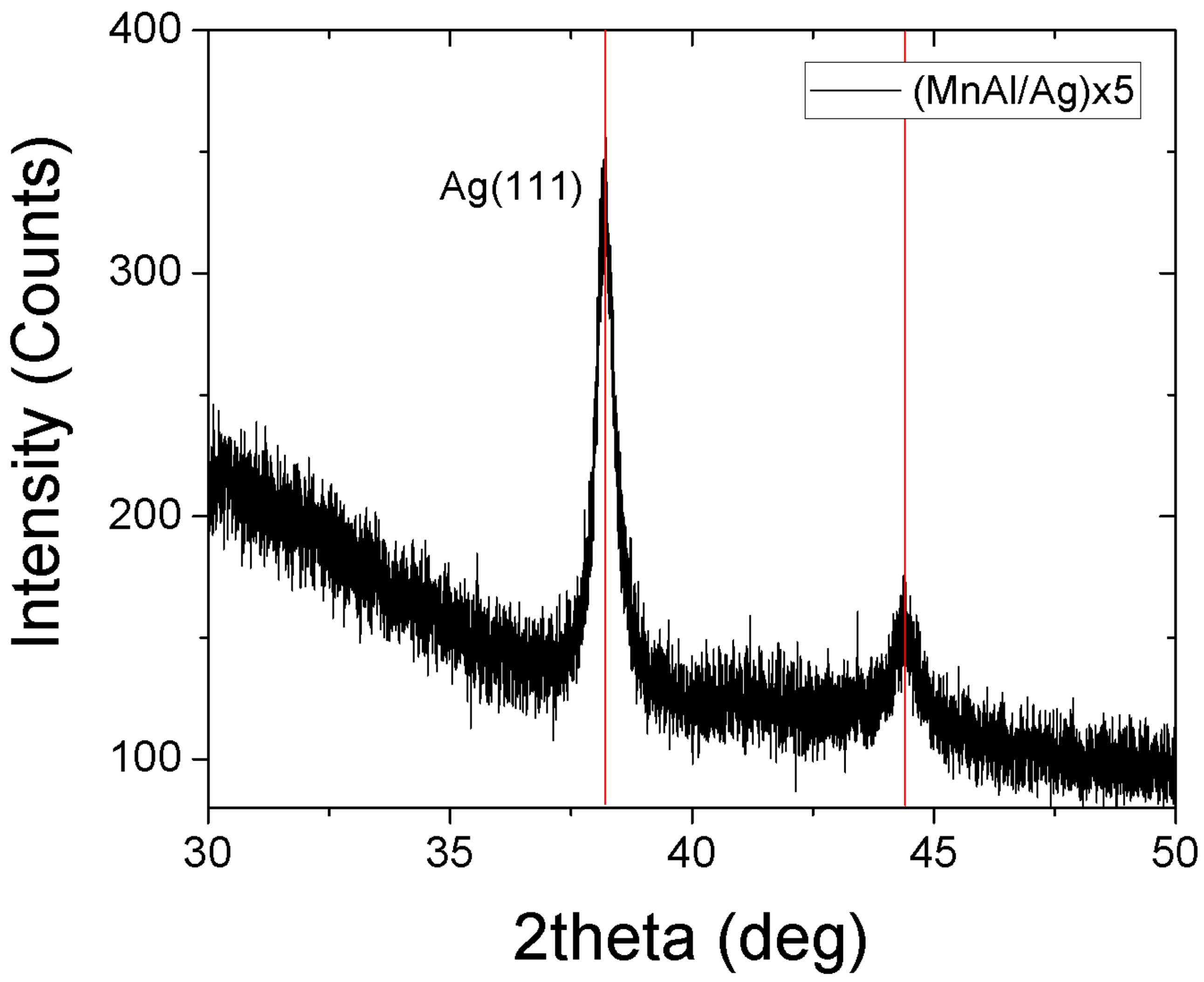
- fcc Ag peak when adding Ag
- Non- τ MnAl

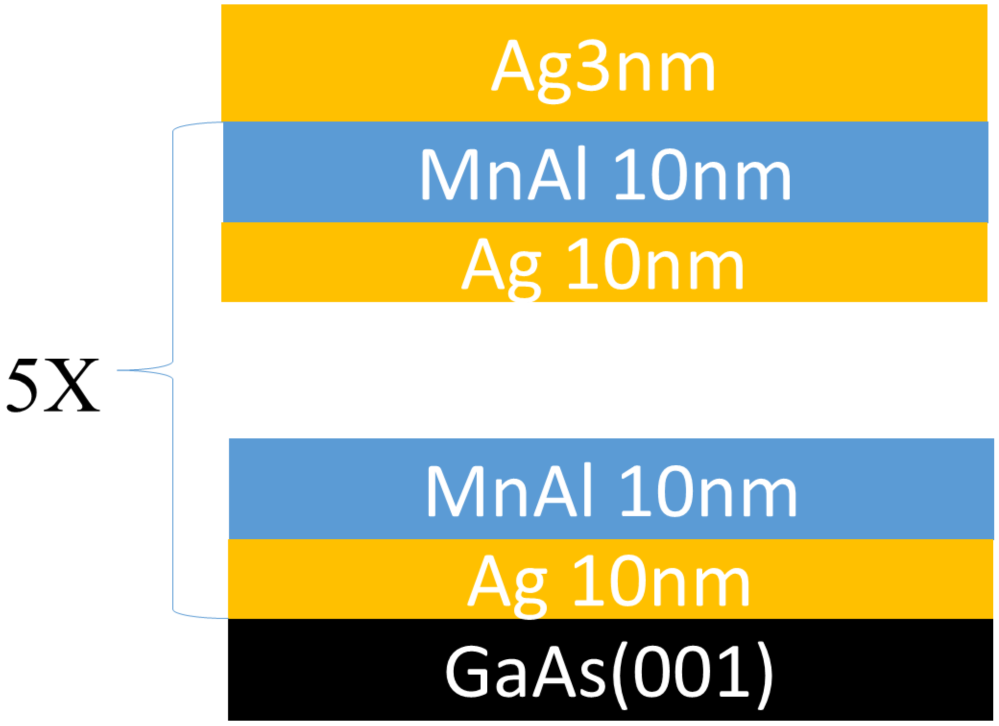
Magnetic characterizations

- MnAl with non- τ phase structure shows quasi-superparamagnetic behavior
- Ag2MnAl shows ferromagnetic behavior, while Mn concentration same as MnAl and Ag5MnAl



Ag/MnAl Multilayered thin films

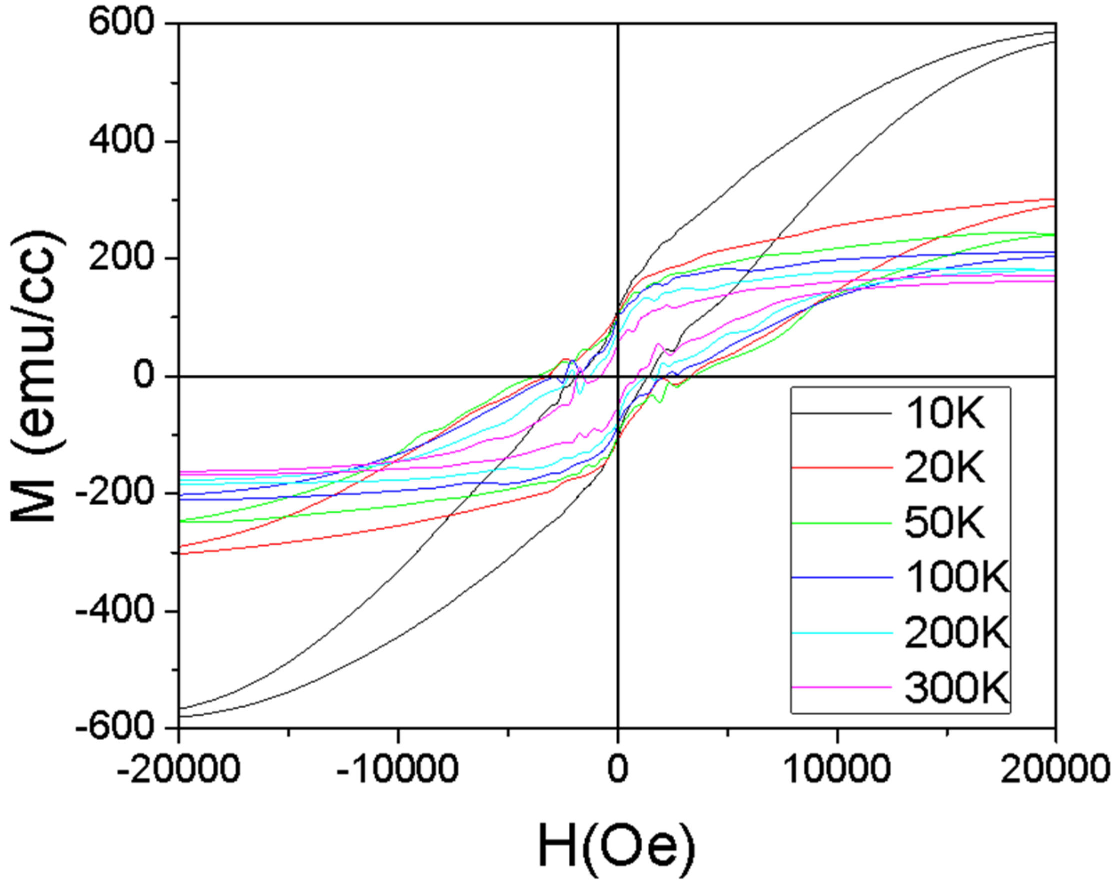




Deposited at 250 Celsius

Magnetic characterizations

- Ferromagnetic phase in multilayered Ag/MnAl thin films.
- Saturation Magnetization increases as temperature decreases.
- Low temperature measurement of multilayered Ag2(MnAl) indicates coexistence of other magnetic configurations.



Future directions

- In summary, our preliminary work suggests that the SPR approach is feasible.
- We plan to frow samples under ND and OAD geometries and compare the angular dependence of photoemission and QE.
- Optimize overlayer CsOx deposition to further improve QE.
- Optimize silmanal thin-film deposition for optimized spin-polarization
- Grow silmanal onto grating surface and verify spin-polarized photemission.