



Upgrade ACME III electron EDM search with a molecular lens

Xing Wu CFP colloquium, Northwestern University, 01/28/2020





Outline

- Motivation & overview
- Molecular properties of ThO & result from ACME II
- Upgrade ACME III with electrostatic lens
 - Good electronic state: Q (${}^{3}\Delta_{2}$) state
 - Robust state preparation with STIRAP
 - Efficient electrostatic focusing of molecule beam
 - Other associated technical upgrades
- Conclusion





EDMs probe TeV scale physics

- New theories predict particles at the TeV energy scale.
- Electron EDM sensitive to coupling with T-violating interactions with particles at the 3-30 TeV scale.





EDM measurement scheme

 $H = -\mu \cdot B - d \cdot E$



EDM measurement scheme

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The ThO $H^{3}_{\uparrow}\Delta_{1}$ state

Sum of $\Omega = \Lambda + \Sigma$ (projection of spin on molecular axis). $\Omega = 1$ and $\Lambda = 2 \rightarrow \Sigma = -1$, i.e. here spin is anti-aligned with Ω



M = -1

()

+1

- High effective field.
- Can be easily polarized.
- Low magnetic noise sensitivity.

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Can reverse the direction of $\vec{\mathcal{E}}_{eff}$ either by reversing:

- The lab electric field, E.
- The internal electric field, N.



ſeff

ρf

 $2D\mathcal{E}$

 $\mu \mathcal{B}_z$

 $\mathcal{N} = +1$

eff

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Can reverse the direction of $\vec{\mathcal{E}}_{\rm eff}$ either by reversing:

- The lab electric field, E.
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Measure EDM energy shift, NE correlated frequency $\omega^{\mathcal{NE}}$.

 $d_e \mathcal{E}_{\text{eff}} = -\hbar \omega^{\mathcal{N}\mathcal{E}}$

Apparatus for 2nd generation ACME



- 2. populate single quantum state
- 3. STIRAP transfer to science state

- 4. spin precession
- 5. Read out precession phase



ACME II final result

- Fastest switch 200kHz: Resolve relative population in two orthogonal states (quantum phase measurement)
- Two ways of reversing electric field interaction with eEDM
- Slower switches to distinguish systematic errors
- ACME II data set: 3 months of EDM data (after \approx 1 year of systematic error searches)

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	(13 +	3.1stat	$\pm 2.6_{\rm syst}$		lable 1 Systen
d.	$= (4.5 \pm$. e - 300			Parameter
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- L		I 📥 I			ω_{ST}^{NE} (via θ_{ST}^{H-C})
100		-VIII		1	$P_{\rm ref}^{N \mathcal{E}}$
					\mathcal{E}^{nr}
80				-	$ \mathcal{C} ^{\mathcal{NE}}$ and $ \mathcal{C} ^{\mathcal{NEB}}$
		Ĺ	I	-	$\omega^{\mathcal{E}}$ (via $\mathcal{B}_{z}^{\mathcal{E}}$)
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				-	Total systematic
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-4	-2	0	2	4	Total uncertainty
	Standar	rd deviations fr	om mean		Values are shown in ura

able $1 \mid$ Systematic shifts for $\omega^{\mathcal{NE}}$ and their statistical uncertainties					
Parameter	Shift	Uncertainty			
$\partial {\cal B}_z/\partial z$ and $\partial {\cal B}_z/\partial y$	7	59			
ω_{ST}^{NE} (via θ_{ST}^{H-C})	0	1			
$P_{\rm ref}^{N\mathcal{E}}$	-	109			
\mathcal{E}^{nr}	-56	140			
$ \mathcal{C} ^{\mathcal{NE}}$ and $ \mathcal{C} ^{\mathcal{NEB}}$	77	125			
$\omega^{\mathcal{E}}$ (via $\mathcal{B}_{z}^{\mathcal{E}}$)	1	1			
Other magnetic-field gradients (4)	-	134			
Non-reversing magnetic field, \mathcal{B}_z^{nr}	-	106			
Transverse magnetic fields, \mathcal{B}_{χ}^{nr} , \mathcal{B}_{y}^{nr}	-	92			
Refinement- and readout-laser detunings	-	76			
$ ilde{\mathcal{N}}$ -correlated laser detuning, $arDelta^{\mathcal{N}}$	-	48			
Total systematic	29	310			
Statistical uncertainty		373			
Total uncertainty		486			

Values are shown in μ rad s⁻¹. All uncertainties are added in quadrature. For $\mathcal{E}_{eff} = 78 \text{ GV cm}^{-1}$, $d_e = 10^{-30}e \text{ cm}$ corresponds to $|\omega^{N\mathcal{E}}| = \mathcal{E}_{eff}d_e/\hbar = 119 \ \mu$ rad s⁻¹.

Apparatus for ACME III



Projected sensitivity gain for ACME III

 $\delta d_e = \frac{1}{2T \mathcal{E}_{eff} \sqrt{N}}$

Already include practical constraints, e.g. the demonstrated state preparation efficiency

Improvement	Signal Gain	EDM Sensitivity Gain	
Increased Precession Time	0.20	2.3	
Electrostatic Lens	15	3.9	
SiPM Detector Upgrade	2.3	1.5	
Timing Jitter Noise Reduction	1	1.7	
Total	7.4	23	

4 important aspects of molecular lens

- Good electronic state for lensing

 Q (³Δ₂) state of ThO
- Robust state preparation

 90% sequential STIRAPs
- Efficient molecular beam focusing

 Hexapole electrostatic lens, x19 times flux enhancement
- Rotational cooling upgrade, etc

 more compact
 cover broader Doppler distribution



Q ($^{3}\Delta_{2}$) state: a new resource for the ACME e⁻EDM search



Made first measurement on the relevant properties of Q state, and showed it is ideal for molecular lens:

linear, $D_0 = 4.1D$

 $g_0 = 2.07 \mu_B$

- ✓ Stark shift:
- ✓ Zeeman shift:
- ✓ Transition strength: $d_{Q-C} = 1.0D$
- ✓ Life time (90% c.l.): τ > 62ms

X. Wu et al, NJP (2020) arXiv:1911.03015

Q ($^{3}\Delta_{2}$) state molecule-frame electric dipole

 Differential Stark-shift measurement of Q—C transition X. Wu et al, NJP (2020) arXiv:1911.03015



Q ($^{3}\Delta_{2}$) state molecule-frame magnetic dipole

• Differential Zeeman-shift measurement of Q—C transition



X. Wu et al, NJP (2020) arXiv:1911.03015

Together with transition with π -polarization

 $g_Q = 2.07(11)$ $g_C = 1.24(6)$ How well can we transfer population into Q state?

STIRAP and Probe Level Scheme in Test Setup:



- ✓ Detect 736nm off-diagonal decay from C—X (v=1), helps a lot to suppress background scattering
- ✓ Both probes excite to EXACTLY the same C state sublevel, so allows direct comparison between population in X and in Q

STIRAP Setup



Efficient population transfer into and out of Q state: 90% STIRAP efficiency



Efficient population transfer into and out of Q state: 90% STIRAP efficiency



Power saturation scan for STIRAP laser (both Pump & Stokes)



STIRAP efficiency vs. 2-photon detuning scan

STIRAP Efficiency

Model Data S [MHz] Stokes (1196nm) detuning $\Delta_{\sf S}$ [MHz] 0.9 25 25 0.8 0.7 D 510kes (1196nm) detuning 20 0.6 0.5 0.4 15 0.3 $\delta_{\text{2}\gamma}=\text{0MHz}$ $\delta_{2\gamma} = \pm 3 \text{MHz}$ 0.2 $\delta_{2\gamma} = \pm 6 \text{MHz}$ 0.1 10 $\delta_{\text{2\gamma}}=\pm 9\text{MHz}$ 0 10 15 20 25 10 15 20 25 Pump (690nm) detuning Δ_{P} [MHz] Pump (690nm) detuning Δ_{P} [MHz]

★ ★ ★ ★ ★ Electrostatic Focusing ★

Hexapole electrostatic lens

- Quadratic E field distribution
- Harmonic potential for states with Linear Starkshift
- ±22kV, 1.5" diameter: 1.8K trap depth, Δv_{tran}=±10m/s capture range







Electrostatic focusing for ThO molecules



0.35

0.2

0.3

0.4

distance between lens and field-plates [m]

9

0.5

0.6

molecules make it.

 Electrostatic lens focuses molecules into the EDM region, giving > x15 gain in signal (including the efficiency of 'double'-STIRAP)

Electrostatic focusing for ThO molecules


Electrostatic focusing for ThO molecules



• Are these numbers / simulation reliable?????

 Simulation method / package very well understood. Validated multiple times before



X Wu, et al, Science 358, 645 (2017) X Wu, et al, ChemPhysChem 17 (22), 3631-3640 (2016)



Electrode diameter = 19mm



Electrode mounted on Macor stand



Electrode diameter = 19mm

Spacing = 9.5mm



Cross section view



Electrostatic lens mounted on 3-axis translational stage

Other technical upgrades

- Rotational-state cooling for x6 broader linewidth, in more compacted space
- Sideband modulation (90 sidebands, spacing = 330kHz)





Conclusion

- Upgrade with electric hexapole lens
 - Ideal state properties in Q state: 4.1D, 2.07 μ_B , >62ms lifetime
 - State preparation efficiency: 90% with STIRAP
 - Electrostatic focusing: x19 times gain in flux
 - Overall gain: > x15 in signal
- Total EDM sensitivity gain: over one order of magnitude

Improvement	Signal Gain	EDM Sensitivity Gain
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ACME Gen II





Pl's (left to right) David DeMille John Doyle Gerald Gabrielse

Graduate students (left to right) **Cris Panda** Cole Meisenhelder Zack Lasner **Daniel Ang** Xing Wu Jonathan Haefner (right upper corner) Adam West Brendon O'Leary Vitaly Andreev Elizabeth Petrik Nick Hutzler

ACME Collaboration

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EDM sensitivity gain vs. interaction length



Not including gains from

- SiPM upgrade: x1.5
- Ex. Noise Suppresion: x1.7

Stark shift of Q—C transition



Zeeman shift of Q—C transition





$$G_Q$$
= 2.061±0.004 μ_B

 G_{C} = 1.222±0.003 μ_{B}



A quick cross check of g_c via X—C transition





- X state has only ~10⁻³ μ_B
- $G_c=1.71\pm0.02 \text{ MHz/G}$,
 - $1.22{\pm}0.02~\mu_{B}$
- Agrees with the value from Q—C transition

STIRAP setup

- Continuous E-field applied from STIRAP to detection region
- Switch between X-C (690nm) and Q-C (1196nm) probing lasers
- Detect 735nm photon from X-C (v=1) decay for both probes



STIRAP scheme for actual Lens



- For test: X |JM>=|00> to C |JM>=|10> to Q |JM>=|20>
- For actual lens setup: we need to go to Q, |JM>=|22>
 - Apply B offset field
 - Use lasers with x-polarization
 - Need twice the laser power

STIRAP scheme for 1st demonstration



Manipulating the phase space distribution



L_{lens}=45cm, D_{lens-FP}=70cm



1- $\sigma \Delta_v$ =1.7565m/s, at detector





Key Question we try to answer

• Do we have enough laser power to saturate double STIRAP between X and Q state of ThO?

Molecular beam property	Before Lens Entrance
Vertical extent	2 cm
Transverse velocity width (FWHM)	13.5m/s (<i>19.6MHz</i> @690nm, <i>11.3MHz</i> @1196nm, <i>8.3MHz</i> for 2-v linewidth

- Previously, showed 90% transfer efficiency each way. Here, demonstrate saturation of STIRAP in Test setup.
- Show that from Test results + modeling: infer we can saturate the power for the actual Molecular Lens



Different Velocity Distributions between Test and Lens, even after Maximizing Collimator Opening

- Test Setup
 - Molecule collimator fully opened, but limited by fluorescence collection (1" ITO window)
 - Gaussian shape, FWHM=9m/s, (5.5MHz in 2-v linewidth)
- Lens entrance
 - Semi-circle shape (verified by trajectory simulation)
 - FWHM=13.5m/s, (8.3MHz in 2-v linewidth)
- Lens exit
 - Trajectory simulation, FWHM = 4.5m/s
 - fully covered by Test
- Only efficiency vs. $V_{\rm z}$ at Lens Input is in question



STIRAP and Probe Level Scheme in Test Setup:



- ✓ Detect 736nm off-diagonal decay from C—X (v=1), helps a lot to suppress background scattering
- ✓ Both probes excite to EXACTLY the same C state sublevel, so allows direct comparison between population in X and in Q

STIRAP Setup



STIRAP Setup



To Measure STIRAP Saturation: Make Sure Probe Beams Saturate Entire Doppler Width

• General strategy: use max available probe power to get most signal from molecules with large v



Lineshape/Power-scan Modeling: Optical Bloch Equations

- Model input:
 - Laser intensity profile; transition dipoles/branching ratios of X-C, Q-C; laser detuning; Doppler distribution; Laser power
- Time integration of Optical Bloch Equation
 - captures dynamics in the transient process, e.g. 50% decay from C to X, J=0
- Modeled detection efficiency vs. v_z
- ✓ High efficiency even at edges of vdistribution, as desired to simulate situation with lens
- Convolute (integrate) with (over) vdistribution gives lineshape (saturation percentage)



Same STIRAP Efficiency at Narrow 200um Beams as Wider Beams, but with Lower Power



A curiosity: asymmetry in STIRAP vs 2-v Detuning

- Both X/Q-probe beams saturate the Doppler distribution: Δv₂=9m/s (FWHM)
- Blue shift 1-photon detuning, to avoid complication from other states/polarizations



 Peak efficiency is asymmetric w.r.t.
2-v resonance







STIRAP Modeling & Corrections

- Hamiltonian: 3-level system, coupled by 2 laser fields. Input parameters all from measurements:
 - Molecule beam size, D_v=5mm
 - transverse Doppler profile, FWHM=9m/s
 - laser beam profile (1/e² d_v \approx 1.5cm, d_x=420um, 200um)
 - Actual $\delta_{1\text{-v}} \And \delta_{2\text{-v}}$ detuning in the scan
 - X-C & Q-C transition strength measured previously



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 - X-C & Q-C transition strength measured previously
- Correction:
 - Higher remaining X population: decay out of 3-level system not captured by the Hamiltonian, but 50% of them is back to X ground state
 - Lower Q population: Q-C optical pumping by Stokes beam imperfection



Correction in STIRAP Modeling, Part 1:



- Correction to Q population
 - $Q_{corr} = Q_{sim}^* \eta$, where constant rescaling factor, $\eta < 1$ (but ≈ 1)
 - Justified when Stokes beam power held constant, with imperfection pumps Q out.
- Correction to X population
 - $X_{corr} = X_{sim} + (1 X_{sim} Q_{sim}) * 0.5 + Q_{sim} * (1 \eta) * 0.5$
 - (1-X_{sim}-Q_{sim}) is the population decay out of the 3-level system via C state
 - $Q_{sim}^*(1-\eta)$ is the amount of Q population pumped out

Verify STIRAP Modeling, Part 1: Population Transfer vs X-C Pump Power



Correction in STIRAP Modeling, Part 2:

- Correction for Stokes (Q-C) power scan
 - $Q_{corr}=Q_{sim}*\eta$, with constant η , NO longer valid
 - Power dependence captured by rate equation of Q-C pumping
 - Solution approximated by simple exponential behavior: Q=Q_{sim}*(η +exp(-Power/P_{sat.})*(1-η)),

where $P_{sat.}$ is the only additional fit parameter, represents 1/e saturation power of Q-C pumping, while η is the same as in Pump (X-C) scan

Correction to X population modified accordingly








Summary for Modeling

- excellent agreement between model and measurement, using '1.5' fitting parameters
- Verified with 2 different laser beam focus size, 3 different values of detuning, across broad range of Pump and Stokes powers
- Demonstrate predictive power with the 200um focus measurement
- Gives confidence on extrapolating to real Lens condition

Differences in Transition Strength: Test vs Lens

- Keep 200um diameter focus, as in Test
- Clebsch-Gordan coefficients² (Transition moment \propto CG factor²)

	Test	Lens	Power required	15 to 40MHz Stark
690 nm	1/3; No E-field mixing	1/3; small E-field mixing	X ~1.25 to <u>1.5 times</u>	scenario for model)
1196 nm	2/15; No E-field mixing	3/15; x1/2 from full E-mixing	X 1.33 times	Only for keeping a
				Unly for keeping a

Test Setup:





B-field along quantization axis can help eliminate issues from 'wrong' polarization

Depends on E-field:

50 to 100V/cm, or

continuous E-field

Proposed Actual STIRAP Setup Geometry



- Field plates dimension (based on field simulation.
 - $L x H \approx 6 cm x 6 cm$
 - Spacing = 3cm
 - Slit 2mm x 40mm Might want narrower slit for more homogeneous E-field

- E-field: 50 to 100V/cm, depending on the matching to Hexapole lens. For maintain Stark splitting >> 100kHz parity splitting of Q-state in region between STIRAP and lens
- Need 2" diameter quarter-wave plate to make correct polarization
- B-field of ~7G is sufficient to detuning the 'wrong' polarization

Projection of saturation power for Lens

Modeling with the actual v_z distribution
 Beam diameter H=3cm, D=200um, at Lens Input, and actual transition
 Including the η=0.9 imperfection factor strength (including parity mixing)



The Entire V_z Distribution at Lens Input Can Be Well Saturated

• We are able to look for optimized $\delta_{2-\nu}$ with modeling



Conclusion: Laser Power Necessary to Saturate STIRAP is Covered by Current TAs

- STIRAP Power requirement
 - Pump (X-C, 690nm) need 120mW x 2 for Lens Input and (less at) Lens Output.
 Covered by 2x 690nm TAs.
 - get 270mW from each TA, 120mW after fiber
 - Toptica claims their 690nm TA will come online (at earliest) the end of the year
 - Stokes (Q-C, 1196nm) need 300mW x2 for Lens Input & Output. Covered by 1x 1196nm TA (900mW after fiber).
- STIRAP Geometry
 - Require STIRAP beam going along quantization axis (through E-field plates, and along Helmholtz coil axis)
 - Space before Lens is compatible with a 30cm Source—Lens distance

Order-of-magnitude estimate

interaction length, 0.4%x5=2%, assuming the same bg pressure in hypothetical Baseline ACME III. 0.4% is from ACME II Scenario **ACME II, direct ACME II, finite** ACME III, overfocus ACME III, finite vacuum scattering) 'head-on' exposure by lens vacuum scattering $(\Phi_0 * 16) * 2\% = 3e7$ $\Phi_0 = 1e8$ $(\Phi_0 * 16) * 0.5\%$ Φ_0 *0.4% =4e5 particle flux density [moleucles/s/cm^2] [only ThO in J=0] [attenuation calib.] =1.6e9*0.5%=8e6 [only ThO in J=0] Coating area 20cm*5cm*2 30cm*28cm*2 100cm*28cm*2 1.3e7 second [10hrs/day, 1 yr of continuous running] **Running time** Sticking probability 100% Number/cm² in 1year 1.3e15 1.3e11 6e11 2e12 6e14 /cm² [4 Angstrom lattice constant] ThO monolayer latt. size No. monolayer per year 2 2e-4 1e-3 3e-3

Attenuation factor is linear with

Scenario	ACME II, direct 'head-on' exposure	ACME II, finite vacuum scattering)	ACME III, overfocus by lens	ACME III, finite vacuum scattering
particle flux density [moleucles/s/cm^2]	Φ ₀ = 1e8 [only ThO in J=0]	Φ ₀ *0.4% =4e5 [attenuation calib.]	(Φ ₀ *16)*0.5% =1.6e9*0.5%=8e6	(Φ ₀ *16)*2%=3e7 [only ThO in J=0]
No. monolayer per year	2	2e-4	1e-3	3e-3
Particle flux density	Underestimated by up to 1 order of magnitude (diff. J, diff. species)	Underestimated by up to 1 order of magnitude (diff. J, diff. species)		Underestimated by up to 1 order of magnitude (diff. J, diff. species)
Attenuation probability		factor of 0.4% overestimated by likely 1~2 orders of magnitude		factor of 2% overestimated by likely 1~2 orders of magnitude
Percent of 'too slow'			factor of 0.5% underestimated by likely 1 order of magnitude	
More realistic No. monolayer per year	2~20	2e-5	<1e-2	3e-4

Looking at the trajectories

• Major difference between good & bad: longitudinal velocities



make into the interaction region

Transverse velocity (v_z) (z is the same as defined in ACME II)

 Cannot differentiate good & bad trajectories in v_z before lens. But they get separated after lens because the bad ones are all slower in v_x and hence spend



Trajectory slope (v z/v x)

• The differentiation gets 'doubly' enhanced by looking at the slope (v_z/v_x).



- ACME II molecule flux density:
 - Phi = [photoelectron rate]/[detection efficiency]/[state prep efficiency]*[rep rate]/[beam cross-section]
 - = [7e5 cnt/shot]/[5%]/[75%]*[50 shot/s]/[10cm^2]
 - = 1e8 molecules/s/cm^2

Interaction region collimator:2.4cm x 2.4cmdetection region: 3.1cm x 3.1cm

- Suppose insert ITO coated surface directly onto molecule beam ('head-on'), assuming 100% sticking probability, continuous running for 10hr/day for 1yr Number density on surface: N = Phi*[1.3e7 sec]=1.3e15 molecules/cm^2
- Typical monolayer size: 10^14/cm^2. ThO lattice constant is know, 4 Angstrom: 6e14 molecules/cm^2

Thus, 2 monolayer for one year continuous running at 100% duty cycle

- ACME III molecule flux density, and the 0.5% overfocused molecules:
 - Lens increase flux by x16 times: Phi'=Phi*16=1.6e9 molecules/s/cm^2
 - 0.5% of the trajectories focused into interaction region gets over-focused onto the last 30cm part of the field plates
 - N' = [0.5%]*Phi'*[1.3e7 sec]*[beam cross-section]/[coated area on ITO]
 = [0.5%]* [1.6e9 /s/cm^2]*[1.3e7 sec]*[10cm^2]/[30cm*28cm*2]
 - = 6e11 molecules/cm^2
 - 6e14 per cm² for ThO monolayer
 - 1e-3 monolayer in 1 yr



- **Baseline analysis:** ACME II ThO monolayer deposition from background scattered molecules (Beer's law of beam attenuation)
 - P=3e-7 Torr interaction region.
 - Attenuation is calibrated to be exp(- [length]/14m*[pressure]/uTorr)
 - For 20cm long (shorter in ACME II), exp(-.2/14*0.3)=99.6%. Thus, .4% attenuation of the beam.
 - Assuming these scattered molecules evenly distributed on 20cm long 5cm wide stripes in all 4 site of the molecule beam
 - Surface density: N''= [.4%]*Phi*[1.3e7 s]*[10cm^2]/[20cm*5cm*4]

= 1.3e11 /cm^2

Phi= 1e8 /s/cm^2, for ACME II

- This is 0.2e-3 monolayer in 1 yr.
- Overfocused trajectories in ACME III: N' = 6e11/cm^2 , x5 of Baseline value
- Both values scale linearly with Phi, so N'/N" ≈ 5 is independent from Phi calibration error

- ACME III, ThO monolayer deposition from background scattered molecules (Beer's law of beam attenuation):
 - X16 larger flux compared to ACME II
 - length factor drops out in the [attenuation factor]/[coated length]
 - Thus, the coated surface density N'''=16*N''= 16*[1.3e11/cm^2]=2e12/cm^2
 - The deposit rate is N'''/[6e14/cm^2]=3e-3 monolayer in 1 yr
 - Overfocused trajectories in ACME III: N' = 6e11/cm^2 = .3* N'''
- It seems the background scattering in ACME III deposit monolayer on ITO surface at x3 higher rate than the overfocused molecule trajectories

To avoid hitting the extended field plates

- '0th order' approximation:
 - Using ideal lens formula, and object has finite size (no aberration, no fuzziness)
 - Magnification=di/do=Si/So



To avoid hitting the extended field plates

- '0th order' approximation:
 - Using ideal lens formula, and object has finite size (no aberration, no fuzziness)
 - Magnification=di/do=Si/So
- Longer field plates → larger image. If take aberration (i.e. 'fuzziness' of the image) into account:
 - \rightarrow smaller signal for a given finite detection volume (we already knew)
 - \rightarrow more likely to hit the field plates



- Lens-to-field-plates distance as short as possible
- bigger field-plates separation, D

