Search for an atomic EDM in hyperpolarized liquid ^{129}Xe

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Permanent EDM of an atom: violates symmetry under P, T, CP

- P violation : Bi, Cs, Tl, …
- CP violation : *K* (1964), *B* (2001).

Atomic EDM search

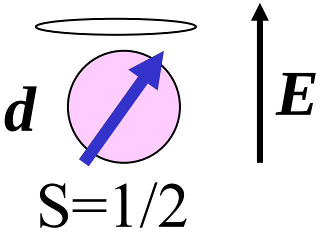
- first evidence of CP violation in eV-level experiments
- table-top test for theories beyond standard model

Published upper limits for diamagnetic atoms

Atom	Year	95% upper bound [e·cm]	Ref.
^{129}Xe	1984	2.3E-26	Vold, Raab, Heckel, Fortson, PRL 52, 2229
	2001	6.6E-27	Rosenberry, Chupp, PRL 86, 22
^{199}Hg	1987	3.4E-26	Lamoreaux et al., PRL 59, 2275
	1993	1.3E-27	Jacobs et al., PRL 71, 3782
	1995	8.7E-28	Jacobs et al. PRA 52, 3521
	2001	2.1E-28	Romalis, Griffith, Jacobs, Fortson, PRL 86, 2505
Rn, Ra	?		

EDM search by precession frequency measurement

$$\omega_{\pm E} = \pm 2\, d\, E / \hbar$$
$$\delta\omega \sim (1/T_2)\,(1/\mathrm{SNR})$$



Motivation for LXe :

- Spin-polarized atomic liquid
  - – Particle density $n \sim 10^{22}$ cm^{-3} (×10^8 rel. to ^{199}Hg)
  - – Dielectric strength $E_{\mathrm{max}} \sim$ 400 kV/cm (×50 rel. to ^{199}Hg)
- Greatly improved statistical sensitivity

EDM search in condensed phase :

- Alignment of dipoles (solid state garnet, B. J. Heidenreich et al. PRL 2005, *also* C-Y Liu, Indiana Univ.)
  - – Transverse coherence not required.
  - – Strong spin-spin interaction in solid phase is ok. (even desired)
- Precession measurement with liquid
  - – Motional average helps, but long range dipolar interaction survives.

✓How much effect on the coherence time & statistical sensitivity?
✓How much systematic effects will be introduced?

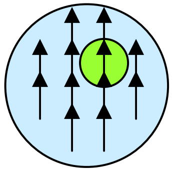
Dipolar interaction

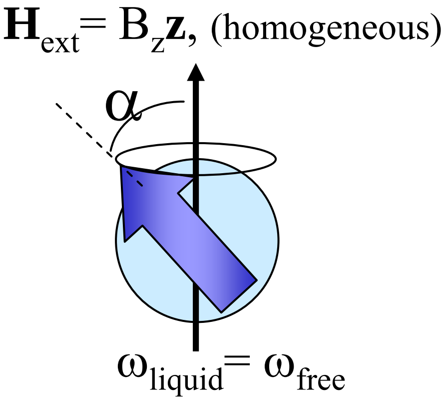
- Spherical cell, uniform magnetization

$$\mathbf{B} = \mathbf{H}+4\pi\mathbf{M}$$
$$= \mathbf{H}_{ext}+\cancel{\mathbf{H}_{pole}}+\cancel{4\pi\mathbf{M}/3}+8\pi\mathbf{M}\cancel{\overline{\delta(\mathbf{r})}}/3$$

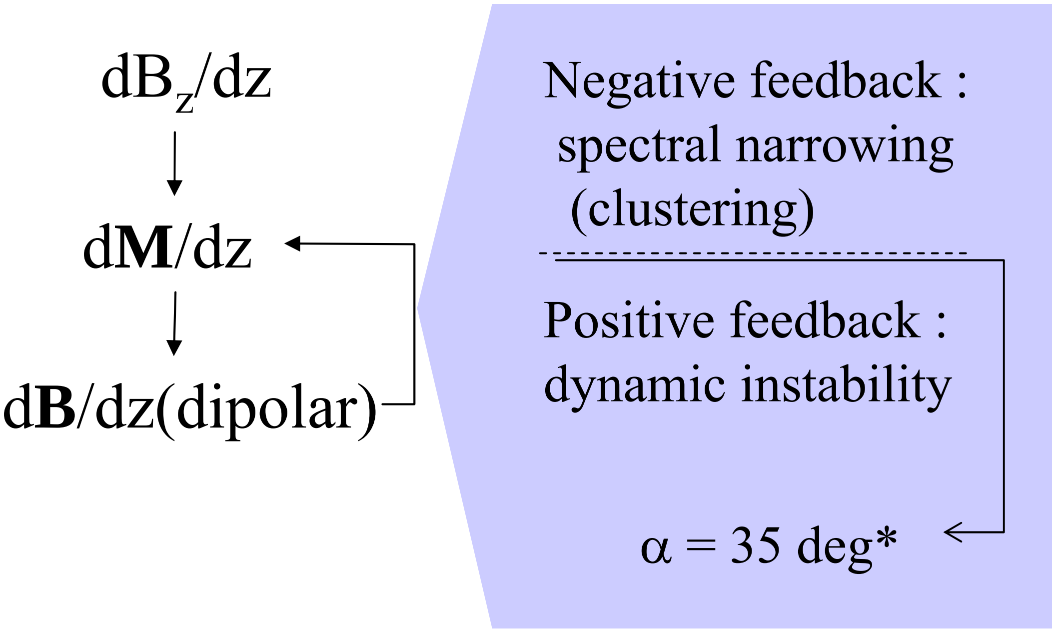
zero for non-overlapping particles

$-4\pi\mathbf{M}/3$ for sphere





- External field gradient, non-uniform magnetization, non-spherical cell
  → nonlinear evolution



dB_z/dz

↓

$d\mathbf{M}/dz$

↓

$d\mathbf{B}/dz$(dipolar)

Negative feedback : spectral narrowing (clustering)

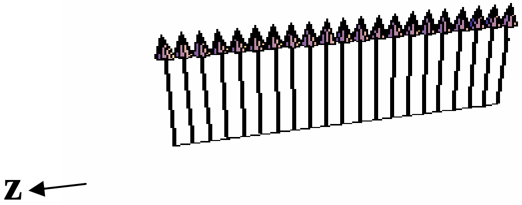
Positive feedback : dynamic instability

α = 35 deg*

* Ledbetter and Romalis, PRL 2002, Ledbetter et al. J. Chem. Phys. 2004, *also*, Jeener, PRL 1999

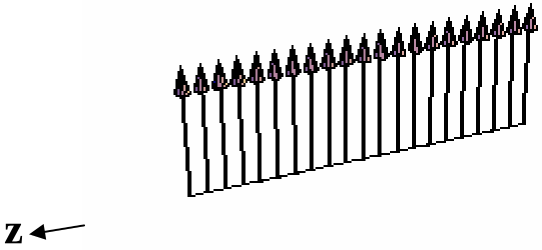
Evolution of transverse magnetization in the rotating frame

free spins,
under gradient
$\Delta B_z = g\ \Delta z$



dephase at a steady rate
$\Delta f = \gamma\ \Delta B/2\pi$

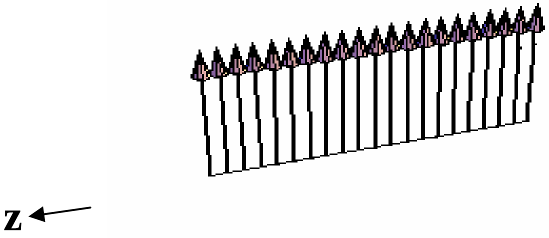
coupled spins,
$\alpha < 35$ deg



oscillating phase gradient

$$\omega = \frac{4\sqrt{2}\pi}{15} M_0 \gamma (3\cos 2\alpha - 1)^{1/2}$$

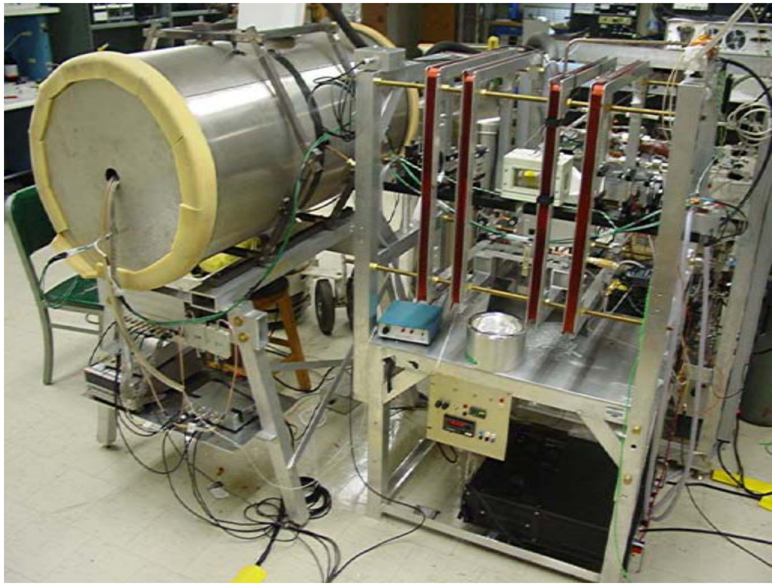
coupled spins,
$\alpha > 35$ deg

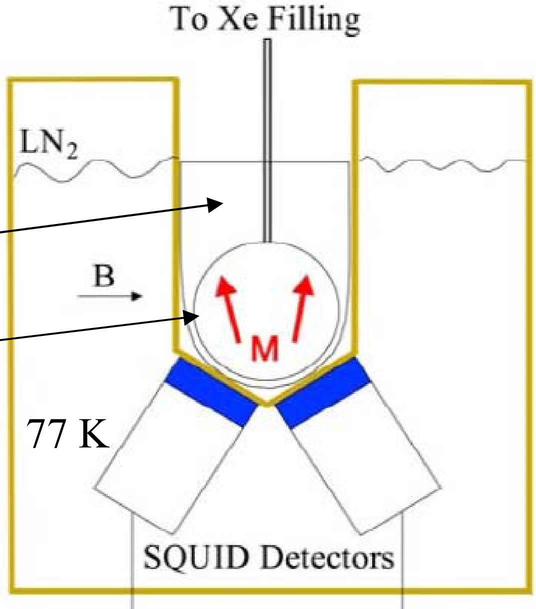


dynamic instability

SQUID detection of Xe precession

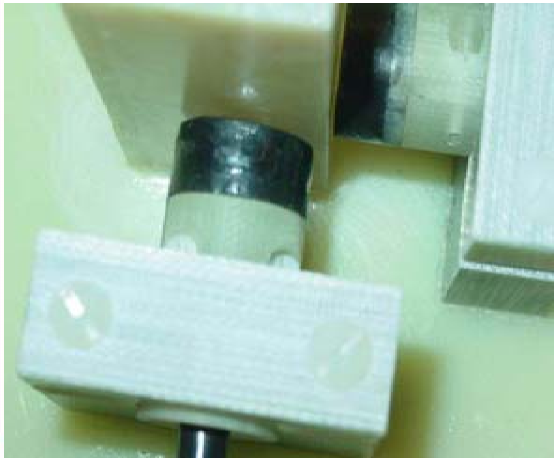
- ^{129}Xe polarization by spin exchange with optically pumped Rb
  - – 1 cm^3(liquid)/20 min, $P = 2 \sim 5$ %
- Five layer mu-metal shield (shielding factor 10^6)
- Detection of precession by a pair of high T_c SQUIDs (Tristan Technologies)
  - – $\delta B \sim 30$ fT/Hz$^{1/2}$, $f_{Xe} \sim 10$ Hz
  - – cell-SQUID separation : 1.5 cm (center-to-center)
  - - Phase difference $\Delta\phi$ in the precession signal picked up by two SQUIDs → phase gradient



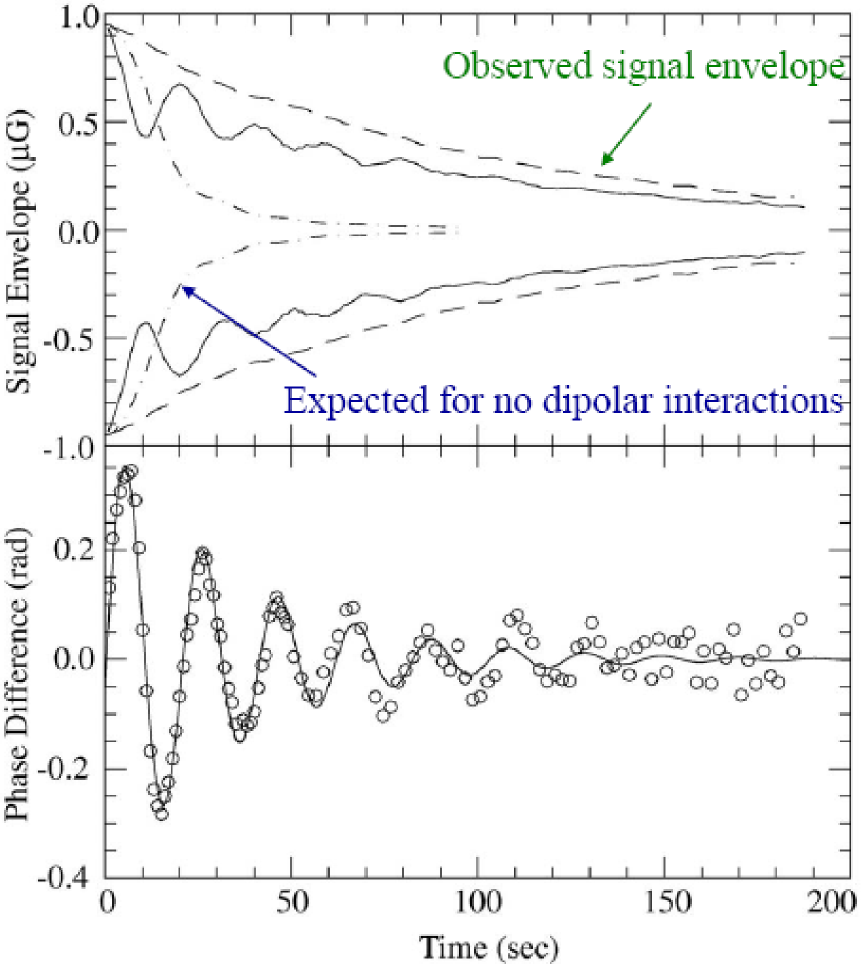


N_2 gas, 160 K

spherical glass cell
1 cm diameter



Tip angle = 3.5 deg



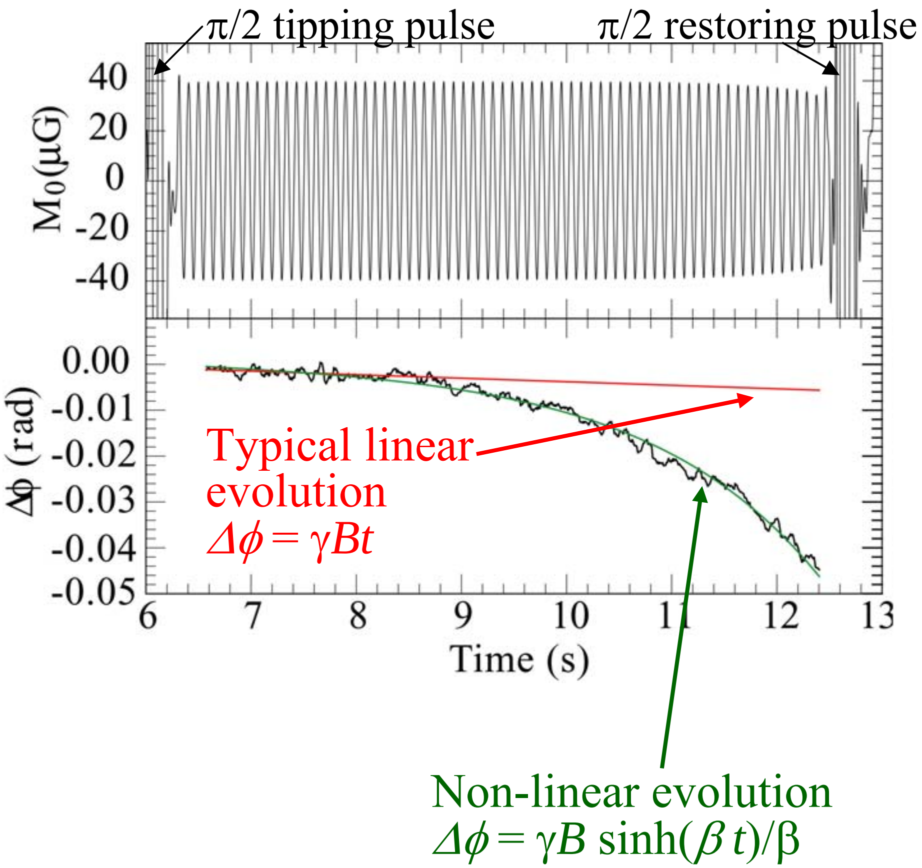
Observed signal envelope

Expected for no dipolar interactions

- T_2^* of >1000 sec obtained without shimming.

Ledbetter and Romalis, PRL (2002).

Tip angle = 90 deg



π/2 tipping pulse

π/2 restoring pulse

Typical linear evolution $\Delta\phi = \gamma B t$

Non-linear evolution $\Delta\phi = \gamma B \sinh(\beta t)/\beta$

Ledbetter, Savukov, Romalis, PRL (2005)

Implication for EDM search

Small tip-angle regime

Precession insensitive to field gradient

→ Use uniform E field

– long coherence time : high SNR

– need to monitor B field drift (co-magnetometer)

First generation EDM experiment

Large tip-angle regime

Exponentially sensitive to field gradient

→ Use E field gradient

– amplification of phase gradient signal by xenon

– need to control ∇B, ∇M

∇T, $\nu(\boldsymbol{r})$,..

Spatially resolved study :
optical detection of nuclear spin polarization

Optical detection of nuclear spin polarization in LXe

Nuclear Spin-induced Optical Rotation (NSOR)

- Faraday rotation enhanced by hyperfine interaction

laser (λ) $\longleftrightarrow$ excited states of electron $\longleftrightarrow$ nuclear spin

- Magnetic field seen by an electron in a nuclear spin-polarized liquid

$$\mathbf{B} = \underset{(=0)}{\mathbf{H}_{ext}} + \underbrace{\mathbf{H}_{pole}(\mathbf{M}) + 4\pi\mathbf{M}/3}_{\text{dist. dipolar field}} + \underbrace{8\pi\boldsymbol{\kappa}\,\mathbf{M}/3}_{\text{contact field,}}$$

$\kappa = 1$ (classical) ⟶

M : nuclear magnetization

nucleus	κ
^{1}H (water)	~1*
^{13}C	4.2§
^{33}S	15§
^{129}Xe	135*

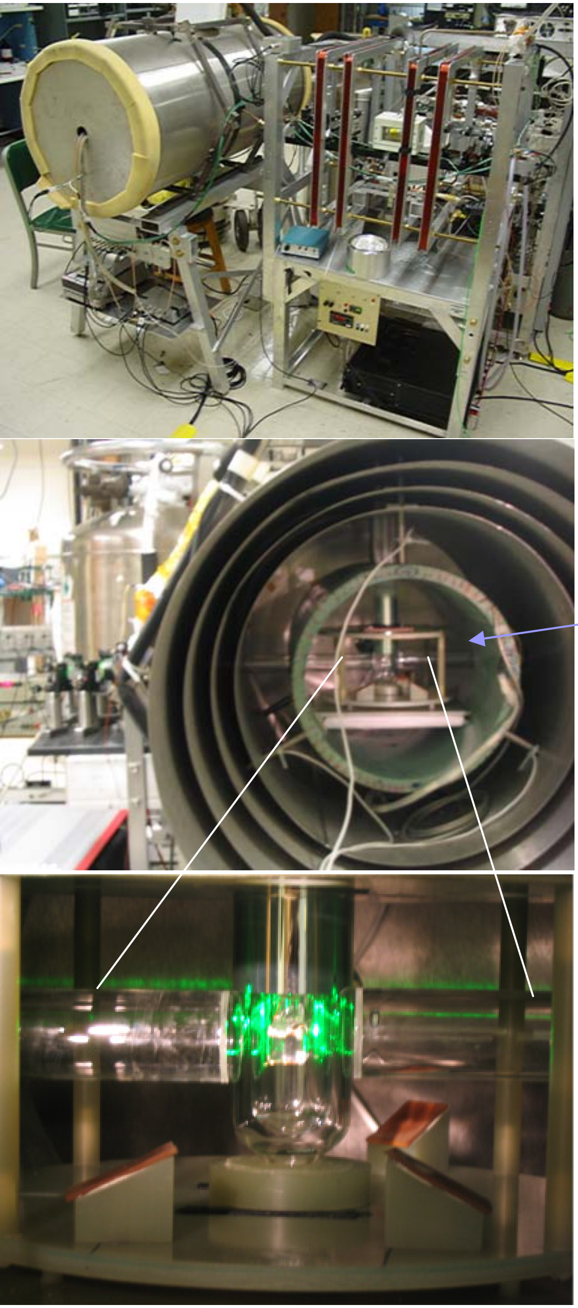
* measured
§ calculated

- Rotation of linear polarization

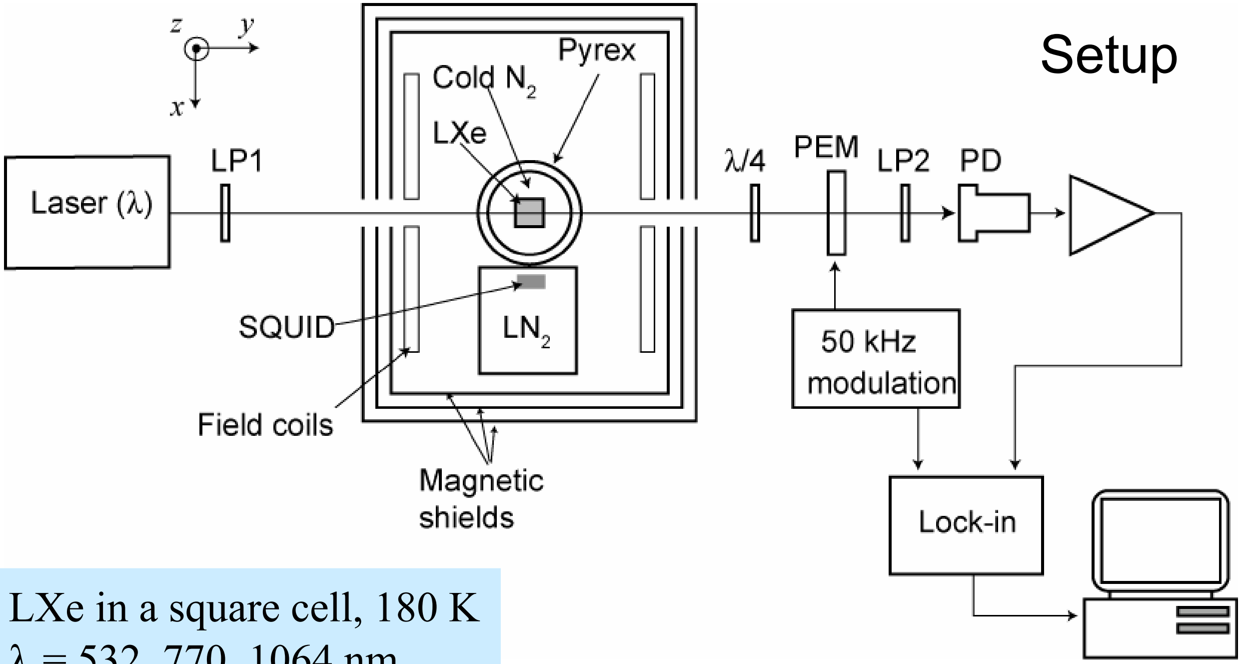
$$\theta_{Faraday} = V(\omega) \cdot l \cdot B$$

$$\theta_{NSOR} = -\frac{4\pi l N r_e c}{n\hbar} \sum_k \frac{f_k a_k \omega^2}{(\omega_k^2 - \omega^2)^2} \langle I_z \rangle$$

N : number density
a_k : hyperfine constant
ω : frequency of laser
n : refractive index
l : path length

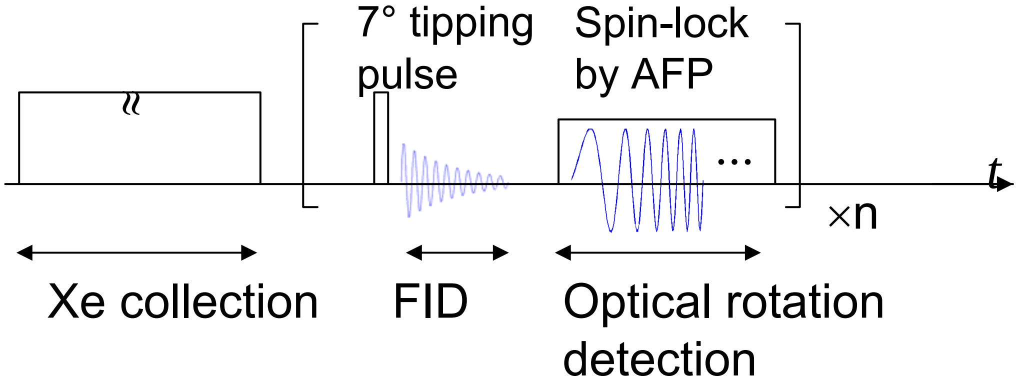


Setup

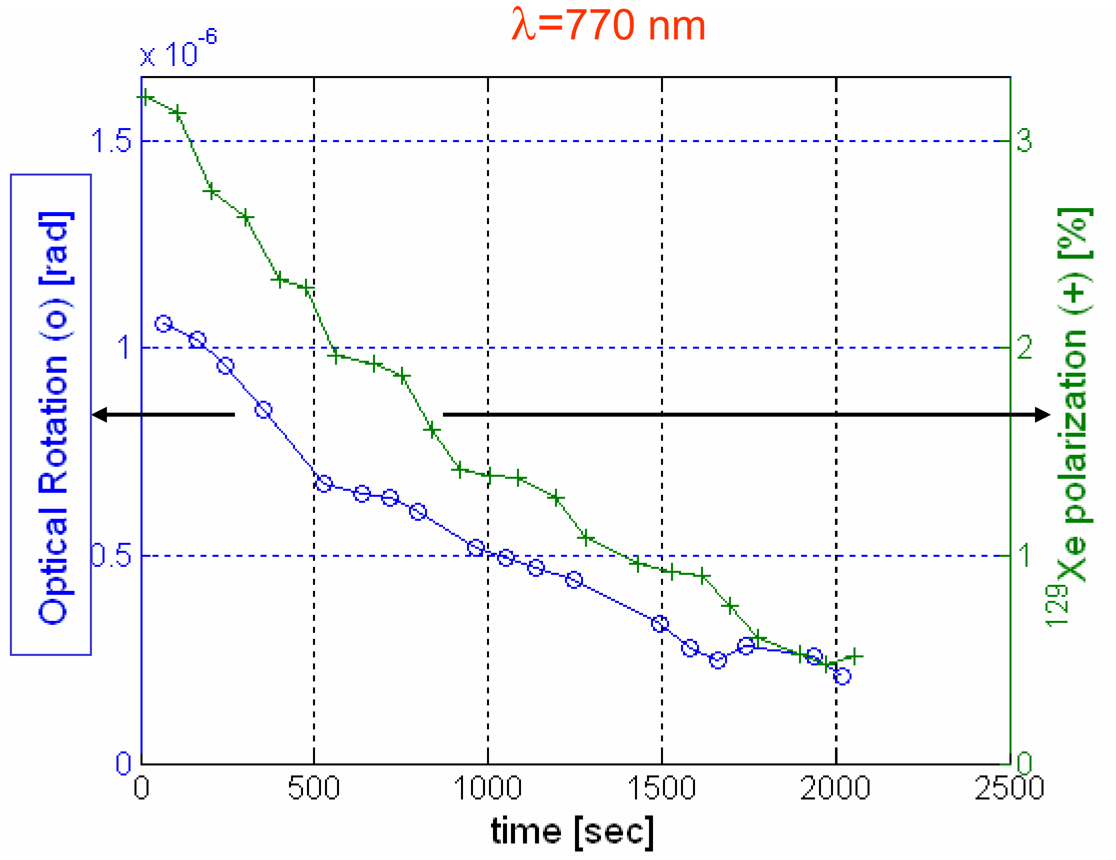


LXe in a square cell, 180 K
λ = 532, 770, 1064 nm

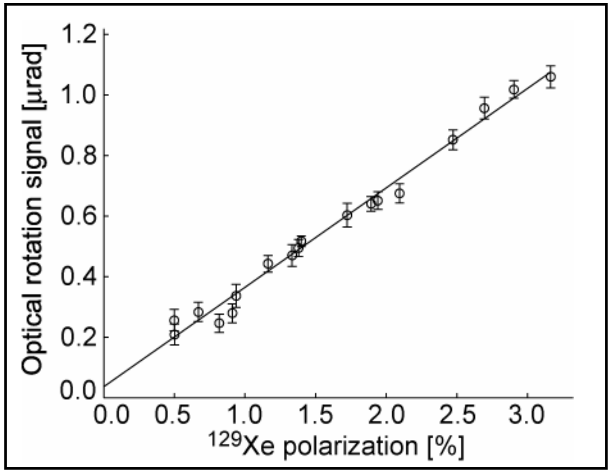
Pulse Sequence



Optically detected ^{129}Xe polarization decay



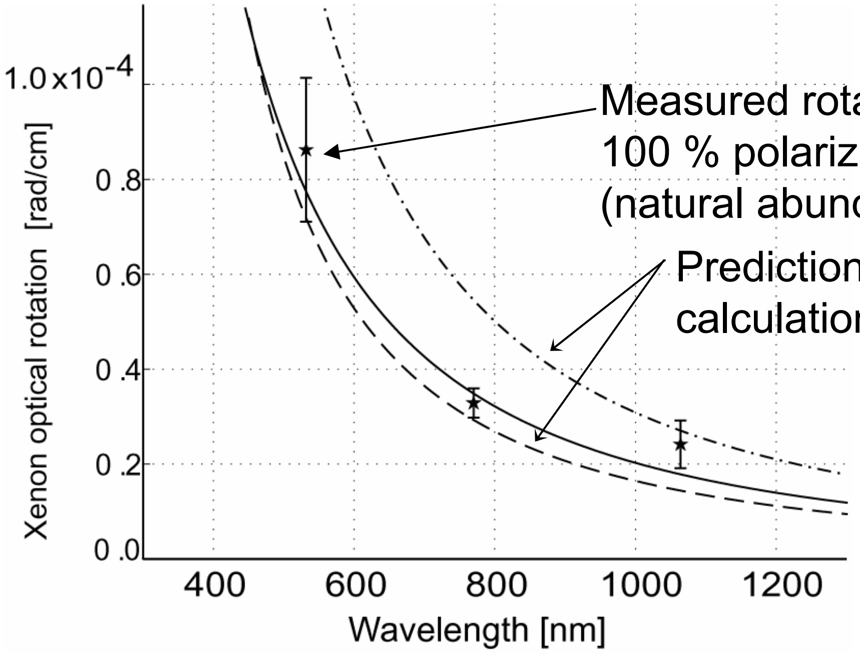
- Decay due to T_1 and small angle tipping pulses



Clear linear relationship between optical rotation and SQUID-detected ^{129}Xe polarization

Savukov, Lee, Romalis, Nature 2006

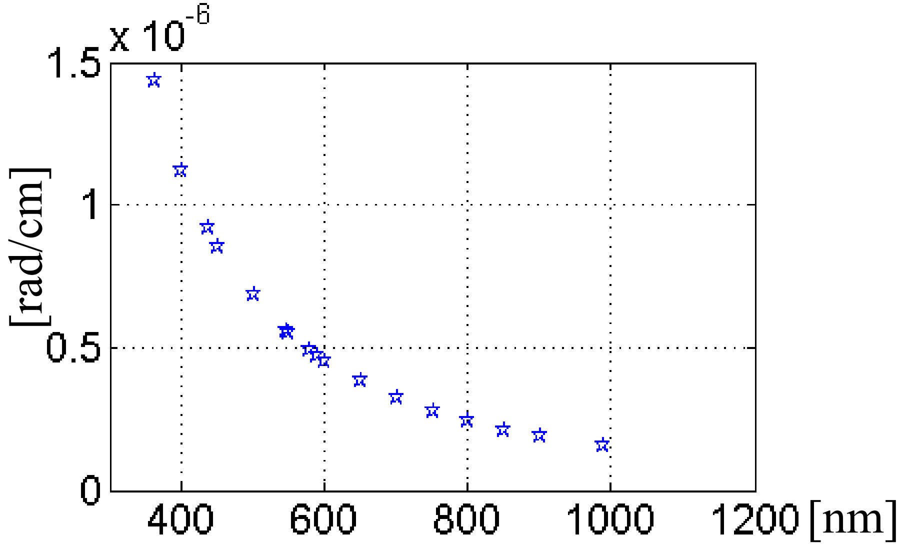
Wavelength dependence of optical rotation in liquid ^{129}Xe



Measured rotation normalized to 100 % polarization of ^{129}Xe (natural abundance)

Predictions from *ab initio* calculation*

* Romero & Vaara, Chem. Phys. Lett. (2004)

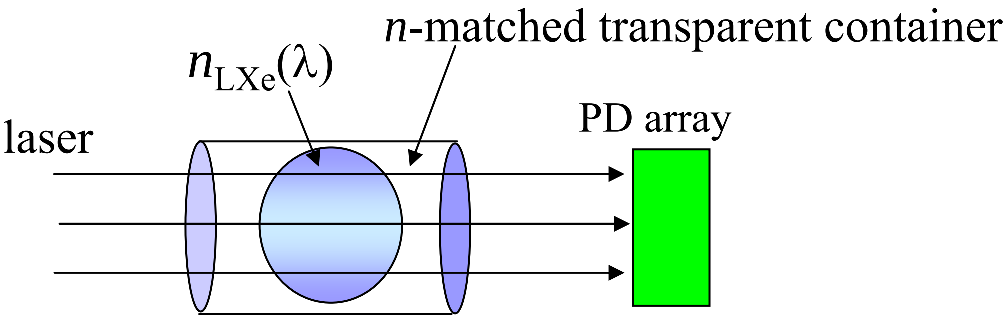


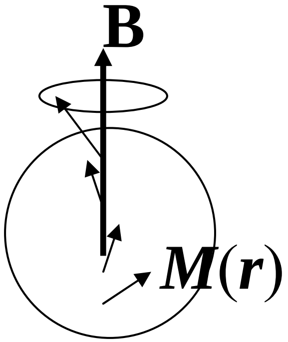
Classical Faraday rotation expected from Verdet constant measurement (Ingersoll et al, J. Opt. Soc. Am. 1956) in gaseous xenon

Future experiment with NSOR

- High resolution NMR imaging of LXe without B-gradient

- study of conditions leading to instability
- directly monitor higher order gradients in ***M***





- Sensitivity enhancement

- current shot-noise-limited SNR ~ $100/\mathrm{s}^{-1/2}$
- UV laser
- cavity/multipass arrangement

→ replace SQUID for EDM signal detection ?

e.g. Properties of LXe @ 178 nm

refractive index	1.7
Rayleigh scattering length	29 cm
absorption length	> 100 cm

LXe EDM Experiment : main features

- Small tip-angle (negative feedback) regime
- Low transition-temperature (T_c) SQUID detection
- SQUID “co-magnetometer”
- Superconducting shield and B field coil

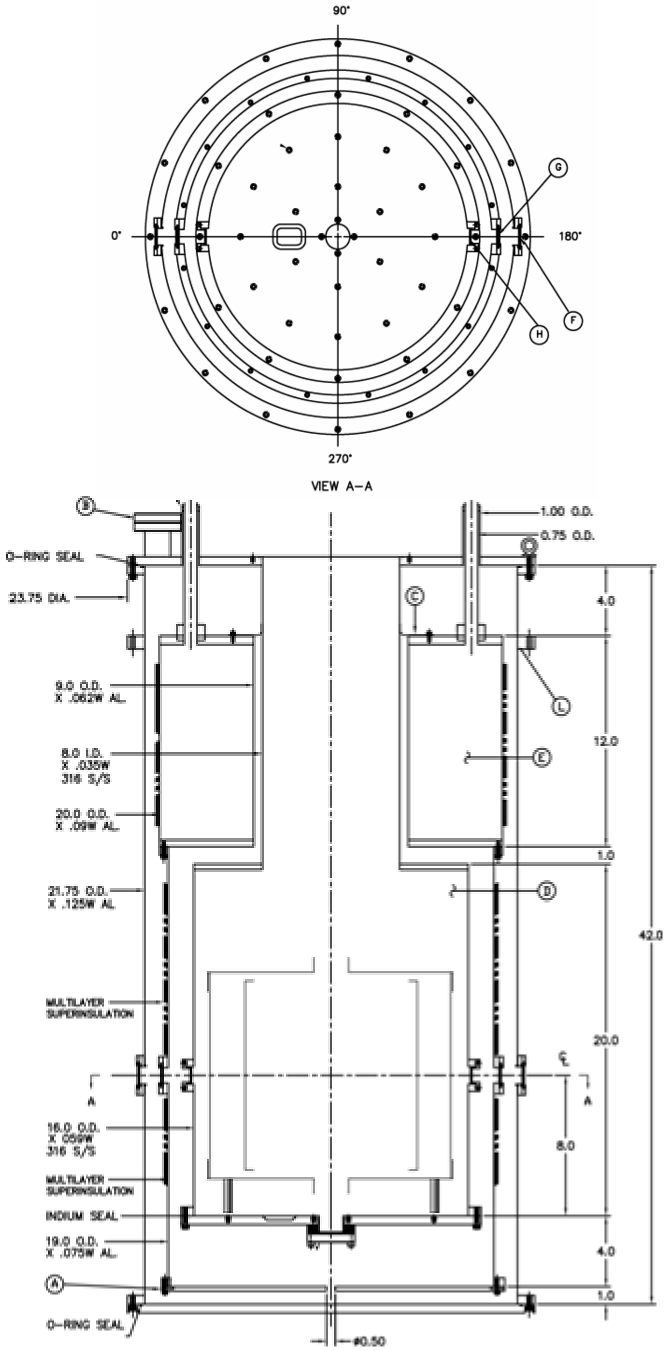
– pushing the limit of NMR frequency and B-field measurement with a SQUID

– sensitivity goal : **$1\ \sigma_d < 10^{-29}$ [e · cm]** (×10 better than ^{199}Hg) in **one day**

→ *equivalents :*

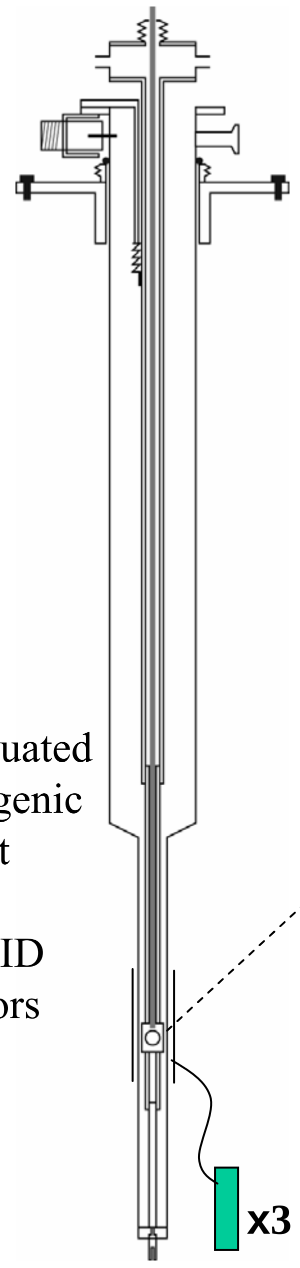
E = 20 kV/cm	E = 80 kV/cm
$\sigma(f) = 0.1$ nHz	$\sigma(f) = 0.4$ nHz
$\sigma(B) = 0.008$ fT	$\sigma(B) = 0.03$ fT
(2.4 fT/Hz$^{1/2}$)	(9 fT/Hz$^{1/2}$)

Setup Overview

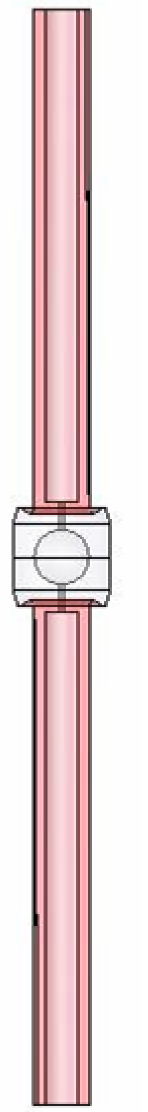


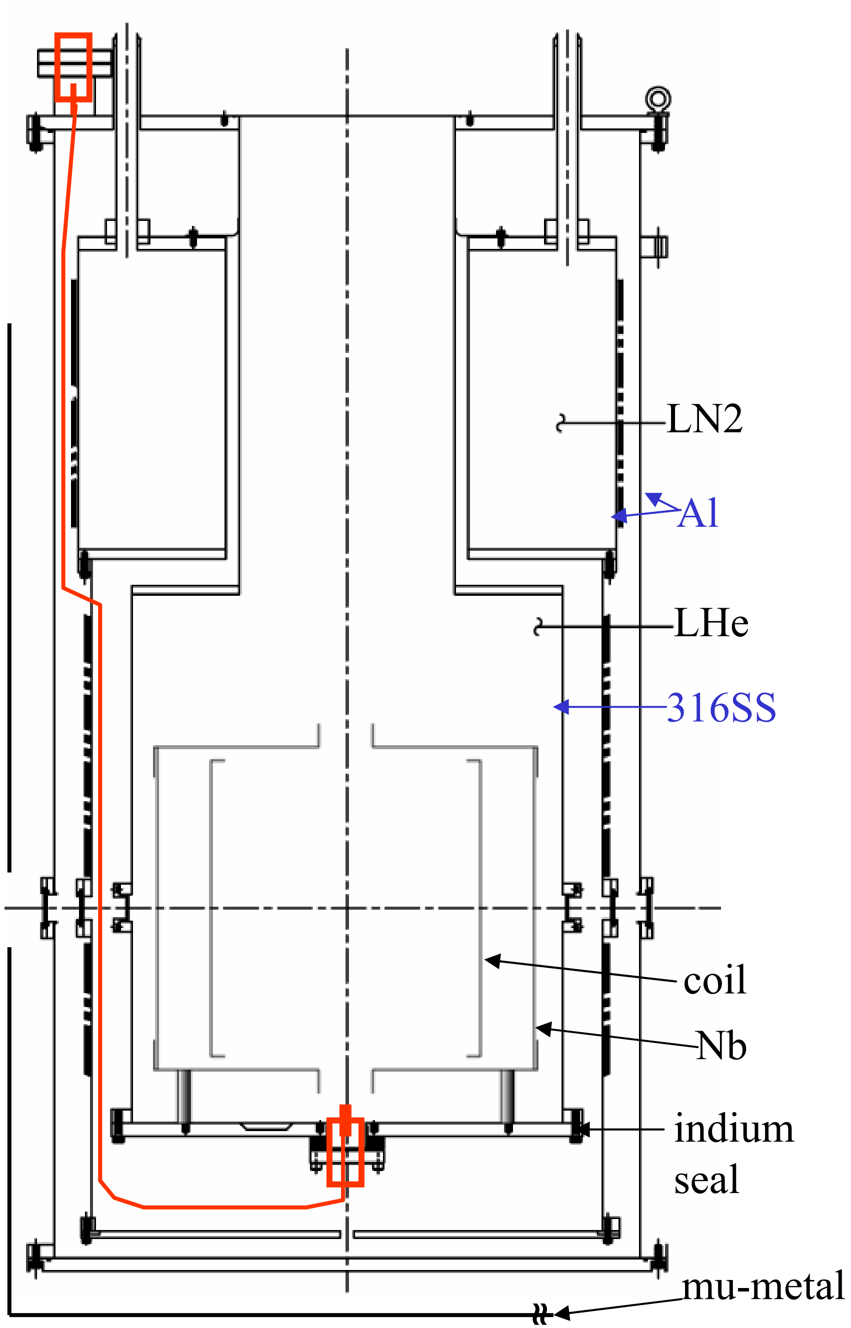
LHe dewar,
Nb shield
and coil

Evacuated
cryogenic
insert
and
SQUID
sensors



sapphire
cell





LHe cryostat

- LN-shielded dewar (unloaded boil-off 6L/day)
- made of aluminum 6061 (outer and LN2 vessel) and 316 stainless steel (LHe vessel)
- demountable bottom flanges for bottom load of large diameter coils and superconducting shield
- Six optical windows made of quartz glass
- HV cable in the vacuum space thermally anchored to the LN2 jacket.

Mu-metal shield

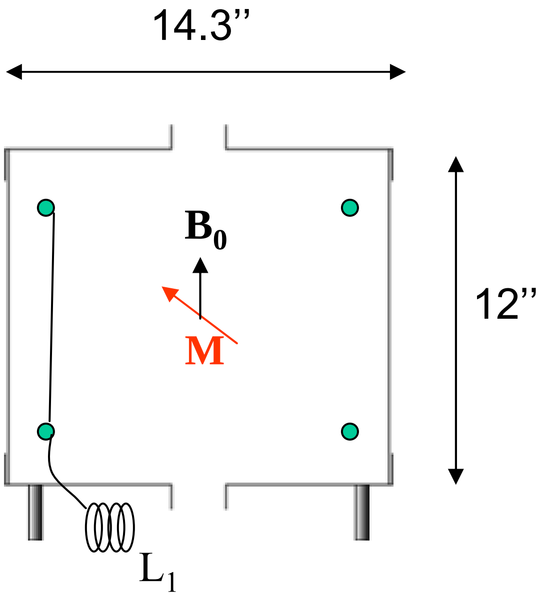
- to shield earth's field when superconducting components are cooled down.

Calculated Johnson noise from structural components

- numerical calculation with actual dimension
- based on electromagnetic power loss and fluctuation-dissipation theorem

Noise from..	temperature	w/o shield [fT/Hz$^{1/2}$]	w/ shield [fT/Hz$^{1/2}$]
HV pin (s.s.1010)	4 K	$4.0\text{x}10^{-2}$	$1.5\text{x}10^{-4}$
LHe vessel (s.s. 316)	4 K	$9.0\text{x}10^{-1}$	$8.4\text{x}10^{-6}$
Thermal shield & LN2 jacket	~80K	$1.8\text{x}10^{1}$	$1.9\text{x}10^{-5}$
Outer vessel	293 K	$4.7\text{x}10^{1}$	$4.7\text{x}10^{-5}$
Mu-metal	293 K	$3.3\text{x}10^{0}$	$1.7\text{x}10^{-6}$

→ Negligible with Nb shield



B_0 field coil :

- Superconducting (Nb-Ti) wire, modified Helmholtz pair
- Homogeneity 1:10^5
- Back-reaction to the dipole suppressed by a large (adjustable) inductor L_1,

$L_{coil} = L_{Helm} + L_1 >> L_{Helm}$

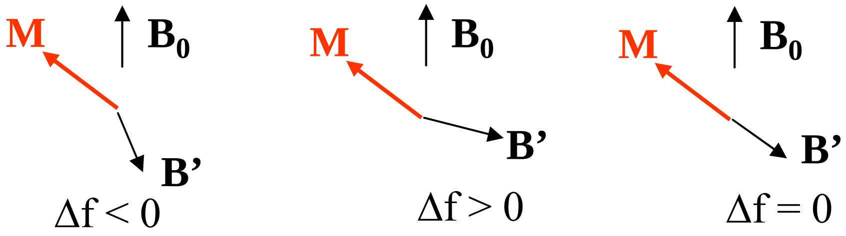
$$B' = M\, b^2/L_{coil}$$

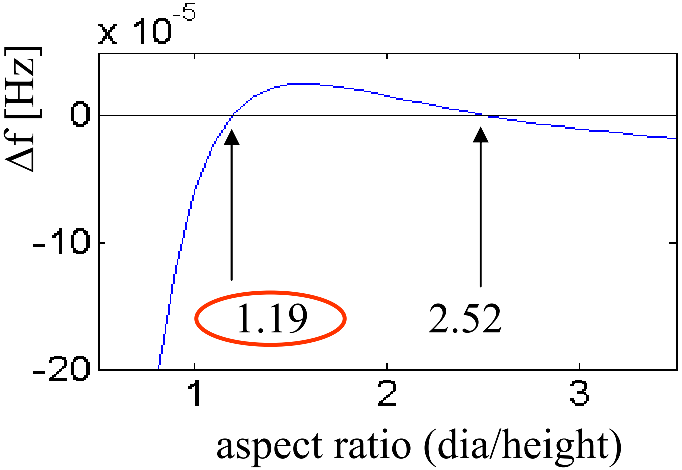
B' : back reaction field
M : dipole moment
b : coil-dipole coupling

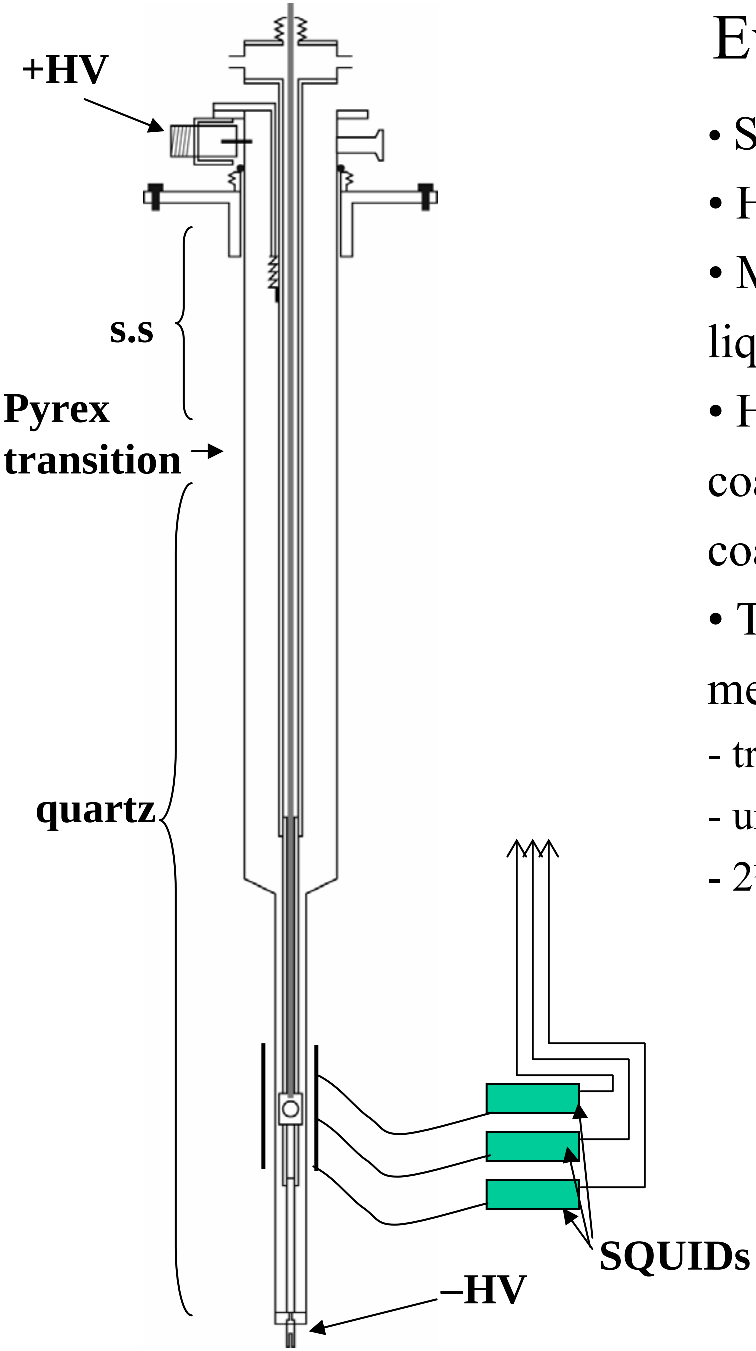
Superconducting Nb shield :

- Six orders of magnitude reduction of environmental field fluctuation, but strongly (dia)magnetic

-A special aspect ratio was chosen where the image field (**B**′) of Nb in response to precessing xenon is always parallel to the xenon itself.



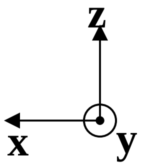




Evacuated LHe insert

- SS-Pyrex-Quartz seal
- HV feedthroughs
- Main function is to provide thermal insulation between liquid xenon and LHe.
- Heat loss : radiation loss suppressed by reflective coating on the inner wall. (Johnson noise suppressed by coating in small ~ 1 mm patches)
- Three sets of SQUID pickup coils configured to measure :

- transverse field (M_x , for Xe precession),
- uniform longitudinal field (B_z , “co-magnetometer”)
- 2nd order gradient longitudinal field (M_z , Xe polarization)

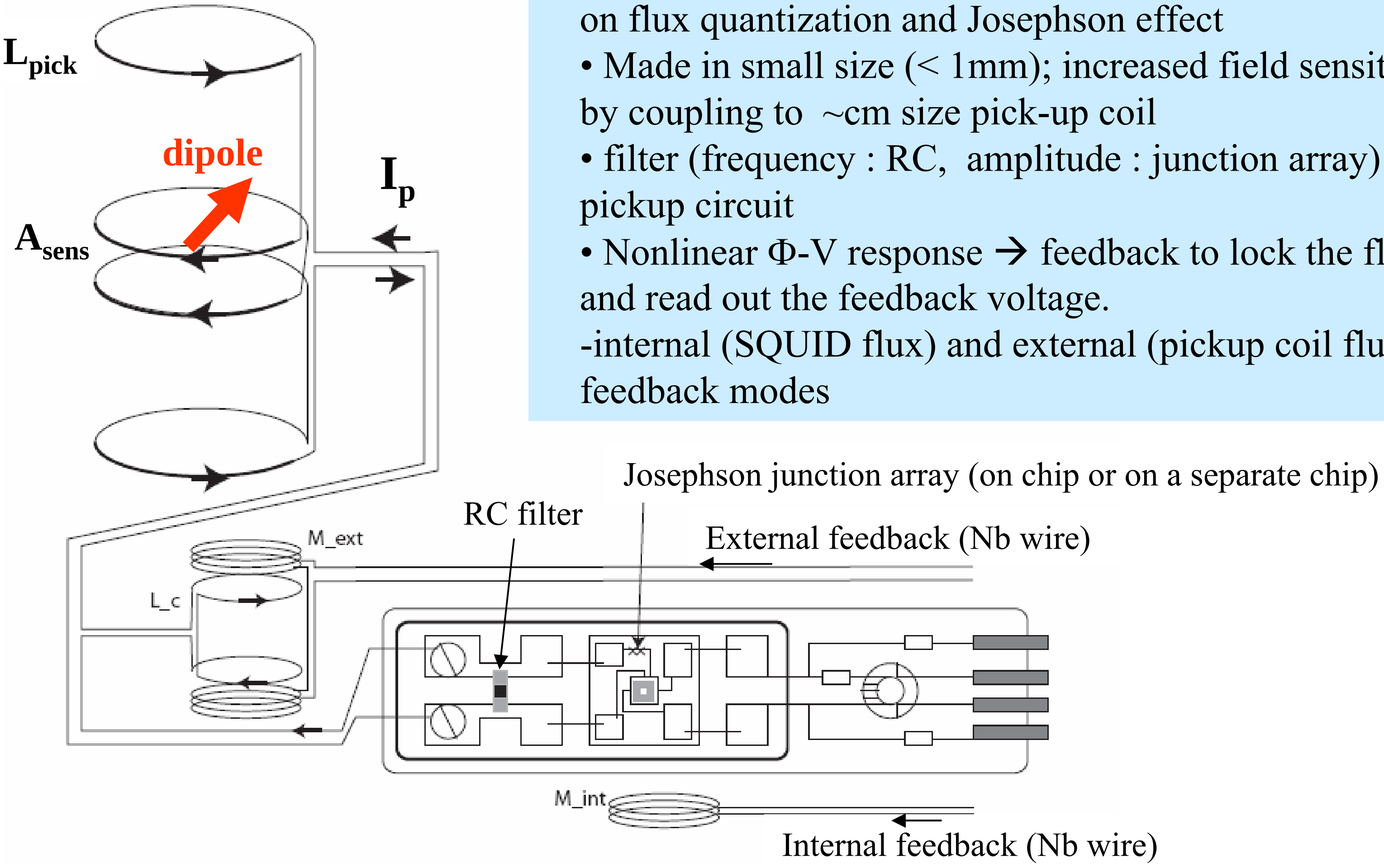


SQUID arrangement

A SQUID Magnetometer

- Detects flux at a level of $\mu\Phi_0$ ($\Phi_0 = 2.07$ fTm^2) based on flux quantization and Josephson effect
- Made in small size (< 1mm); increased field sensitivity by coupling to ~cm size pick-up coil
- filter (frequency : RC, amplitude : junction array) in the pickup circuit
- Nonlinear Φ-V response → feedback to lock the flux and read out the feedback voltage.

-internal (SQUID flux) and external (pickup coil flux) feedback modes



Internal feedback

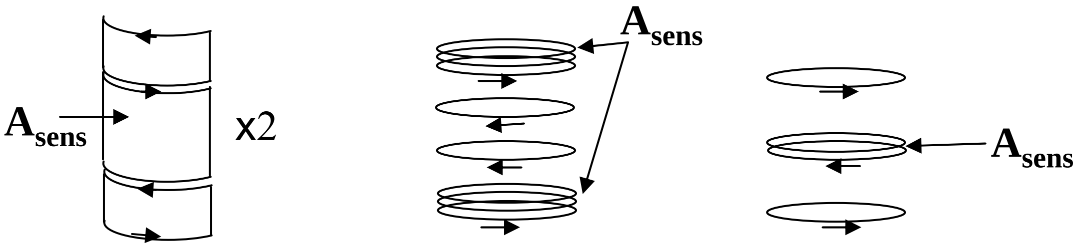
$I_{p,int} = B\,A_{sens} / (L_{pick} + L_{input})$

External feedback

$I_p \approx I_{p,int}$ / (flux loop gain)

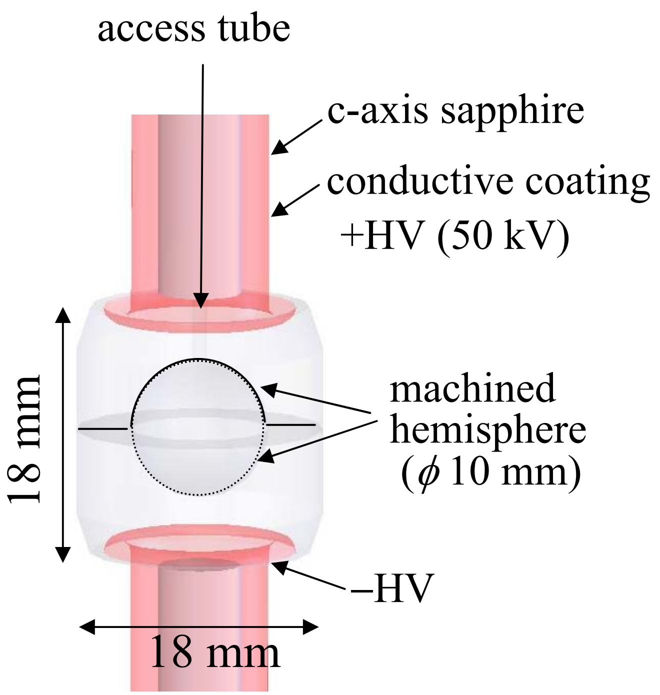
SQUID parameters

dia. = 1 in.



SQUID sensitive to:	M_x	B_z	M_z
Pickup coil geometry	saddle coil	3−1−1+3 circular loops	1−2+1 circular loops
A_{sens}	1620 mm^2	1960 mm^2	980 mm^2
L_{pickup}	0.48 μH	1.6 μH	0.47 μH
A_{eff} for inductance matched SQUID	17 mm^2	11 mm^2	10 mm^2
δB_{sens} for $\delta\Phi = 2\ \mu\Phi_0/Hz^{1/2}$	0.24 $fT/Hz^{1/2}$	0.38 $fT/Hz^{1/2}$	0.42 $fT/Hz^{1/2}$

Sapphire cavity as a LXe cell


access tube

c-axis sapphire

conductive coating

+HV (50 kV)

machined hemisphere (ϕ 10 mm)

–HV

18 mm

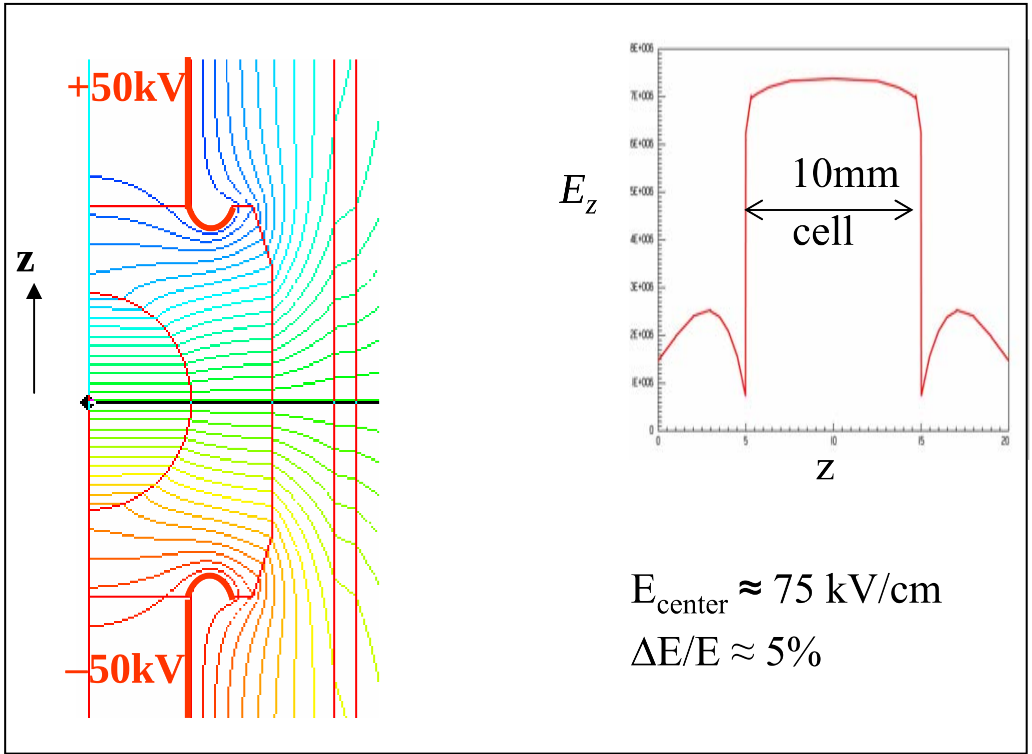
18 mm

Advantages of sapphire (Al_2O_3)

- High thermal conductivity: $\kappa = 70\ Wm^{-1}K^{-1}$ @ 150 K
- High dielectric strength : 480 kV/cm @ R.T.
- Electrical resistivity is large at low temperature.

>10^{17} ←	10^{16}	10^{11}	10^{6} Ωcm
160 K ←	298K	873K	1273K

E field calculation



$E_{center} \approx$ 75 kV/cm

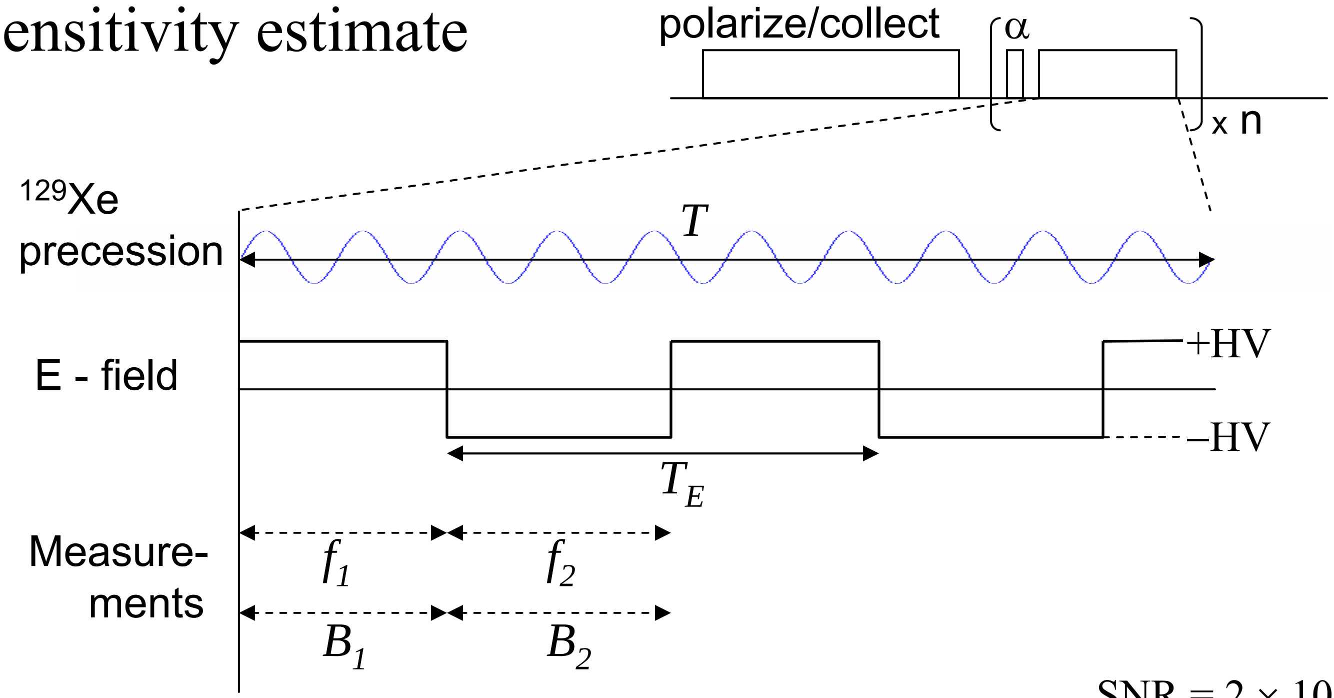
$\Delta E/E \approx 5\%$

- Field amplification inside a dielectric cavity

$E/E_0 = 3/(\varepsilon^{-1}+2) \approx 1.4$

ε(sapphire, ||) = 10

Sensitivity estimate



$$h(f_1 - f_2) = 2\mu_{Xe}(B_1 - B_2) + 4d_{Xe}E$$

$$\sigma^2(d_{Xe}) = \left(\frac{h}{4E}\cdot\sqrt{2}\cdot\underline{\sigma(f_1)}\right)^2 + \left(\frac{\mu_{Xe}}{2E}\cdot\underline{\sigma(B_1 - B_2)}\right)^2$$

$$\begin{cases} \sigma(f) \propto \dfrac{1}{(SNR\_per\_\sqrt{s})}\dfrac{1}{T_E\sqrt{T}} \\ \sigma(B) \propto \delta B(f_{corner})\sqrt{\dfrac{T_E}{T}} \quad \text{(1/}f\text{ noise dominated)} \end{cases}$$

SNR = $2 \times 10^6 / \mathrm{s}^{1/2}$
(natural abundance xenon, 5 % polarization, α = 2 deg),
δB = 0.5 fT/Hz$^{1/2}$ at f_{corner} = 1 Hz ,
T_{E} = 20 s ,
E = 75 kV/cm

→ For one-day average,
$\sigma_d = 3 \times 10^{-30}$ e cm

False positives

$\Delta f \propto E$

- **vxE**
- low temperature, condensed sample
- convection → suppressed by positive (upward) temperature gradient
- **leakage current**
- monitored by Bz and Mz SQUIDs (analogous to 4-cell Hg experiment)
- Additional SQUID for current measurement in the conduction path possible.
- no gas, cell surface in vacuum, high purity cell construction
- **charging current, magnetized impurity**

$\Delta f \propto E^2$

- electrostriction of sapphire → cell deformation
$\delta x/x \sim 0.3$ ppm (r.t), **significant for E reversal asymmetry ~0.1 %**

- non-linear dielectric effect (density change by E) of LXe
$\delta n/n < 8$ ppm, **suppressed by plugged cell**

- cell motion, in the image field and gradient field

→ $\delta(\Delta f)/d(E^2)$ can be directly measured.

Conclusion

- Developed theoretical/numerical/experimental tools to study magnetization evolution of hyperpolarized liquid xenon under the effect of strong dipolar interaction
- Analysis for seamless integration of superconducting components (minimize back-reaction)
- Experimental design to take advantage of high statistical sensitivity provided by LXe
- Physical construction of the setup will start upon receipt of the cryostat (Janis Research) and Nb SQUIDs (ez SQUID, Germany).

→ *First competitive atomic EDM measurement from condensed-phase sample is expected.*