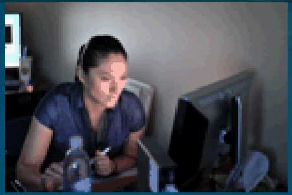
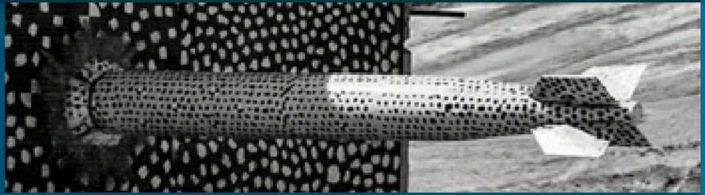
SAND2019-9303PE

Earth's-Field NMR for the Detection and Characterization of Shielded Threats

7 August 2019





PRESENTED BY

Kyle Polack and Peter Marleau

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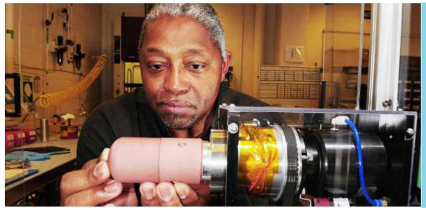
Sandia National Laboratories

- Sandia National Laboratories (SNL) is a multi-faceted national security laboratory responsible for developing technologies to ensure global peace
- SNL is a federally funded research and development center managed by National Technology and Engineering Solutions of Sandia, LLC for the Department of Energy
- SNL's principal sites are in Albuquerque, NM and Livermore, CA, but we also have activities at
  - Kuai Test Facility – Kuai, HI
  - Waste Isolation Pilot Plant – Carlsbad, NM
  - Weapons Evaluation Test Laboratory at the Pantex Plant – Amarillo, TX
  - Tonopah Test Range – Tonopah, NV
- Key Mission Areas
  - Nuclear Deterrence
  - Defense Nuclear Nonproliferation and Global Security
  - National Security Programs
  - Energy and Homeland Security
  - Advanced Science and Technology

SNL National Security Mission Areas

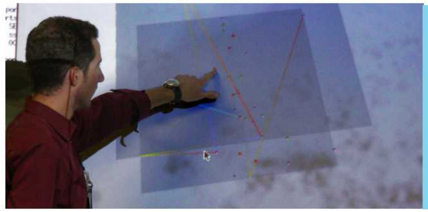


Nuclear Deterrence

Defense Nuclear Nonproliferation

National Security Programs

Energy & Homeland Security



Advanced Science & Technology

[illegible] of life.

Nuclear Proliferation

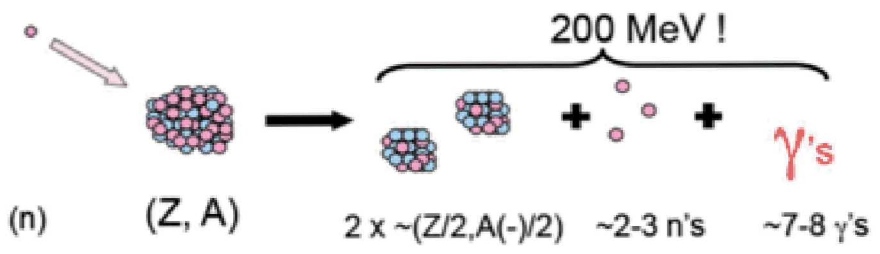
- Nuclear proliferation is the pathway that leads from peaceful use of nuclear power to weaponization of nuclear material and the development of a deliverable nuclear weapon
  - Horizontal proliferation is the spread of nuclear capabilities to new state or non-state actors
  - Vertical proliferation involves existing nuclear weapons states increasing their nuclear capabilities
- Special nuclear material (SNM) is required to make a nuclear weapon
- SNM includes
  - Plutonium
  - Uranium enriched in ^{233}U or ^{235}U

Refined uranium ore, known as "yellowcake"
Photo credit iaea.org

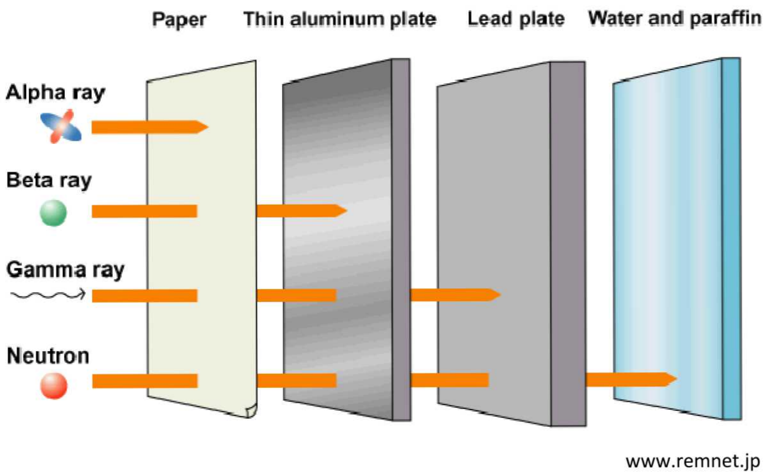
Titan II Missile
Photo credit: titanmisslemuseum.org

Detection of SNM

- What does it look like?
  - Many different forms (e.g. metals, oxides) and colors
  - Could be concealed (by itself or in a weapon)
- •SNM emits ionizing radiation
  - Sensitive and specific signature
  - Only neutral particles useful in most cases
    - Gammas
      - Emitted as decay lines and fission distributions
      - Interact preferentially with high-Z materials (e.g. lead)
      - Energy spectra can be used to determine isotopic content of SNM
    - Neutrons
      - Emitted through fission (spontaneous, self-induced, and externally induced)
      - Certain compositions, such as oxides, will also emit neutrons from (α,n) reactions
      - Interact preferentially with low-Z materials (e.g. hydrogenous materials)
      - Fission energy neutrons are a key signature of SNM
      - Correlated particle analysis can be used to determine SNM properties (e.g. fission rate, multiplication, (α,n) component)
- Systems to improve the ability to **detect**, **localize**, and **characterize** SNM are paramount in the non-proliferation and nuclear security mission space



The nuclear fission process



Radiation penetration through materials

Radiation and Nuclear Detection Systems

Department Overview

- Specialize in designing detection systems for specific applications, radiation signatures, environments, and user requirements

- Performs R&D of ionizing radiation and rare signature detection systems to address broad nuclear security and non-proliferation needs
  - Nuclear proliferation detection
  - International safeguards
  - Nuclear arms control treaty verification
  - Radiological emergency response
  - Counter terrorism

- Areas of significant experience and expertise include
  - Fission-energy neutron detection
  - Standoff detection and localization of radiological materials
  - Antineutrino and coherent neutrino scattering detection
  - Rare event and weak source detection techniques
  - High-resolution imaging
  - Particle identification and discrimination
  - Active interrogation and neutron generators

Arms control treaty verification

Cargo screening

Safeguard s

Emergency response

Radiation Detector Development at SNL

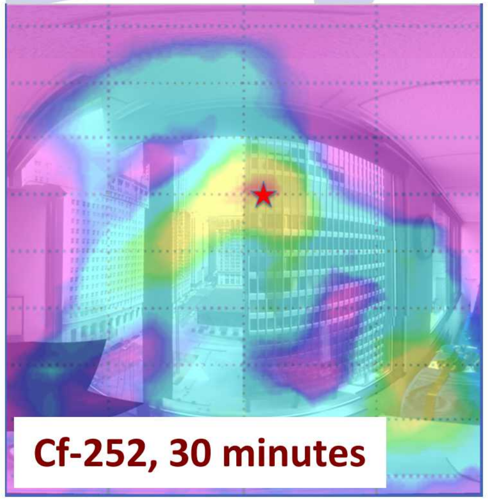
- Neutron scatter cameras
  - Generate low-resolution images
  - Maintain energy information

NSC

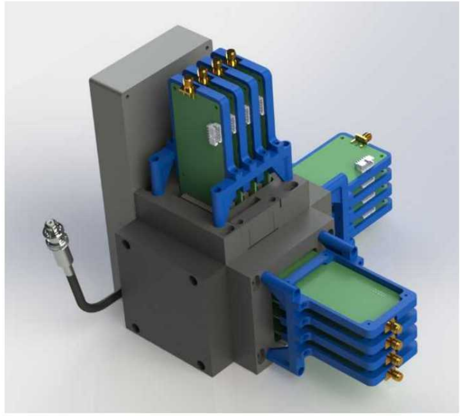
- 2 planes of 16 5” diameter liquid scintillators (front plane 2” thick, rear plane 5” thick)
- Variable planar gap

MINER

- 16 3”x3” liquid scintillators
- Compact
- Battery operable
- More uniform field of view



High-rise to high-rise source localization



SVSC

- Single scintillating volume
- More compact

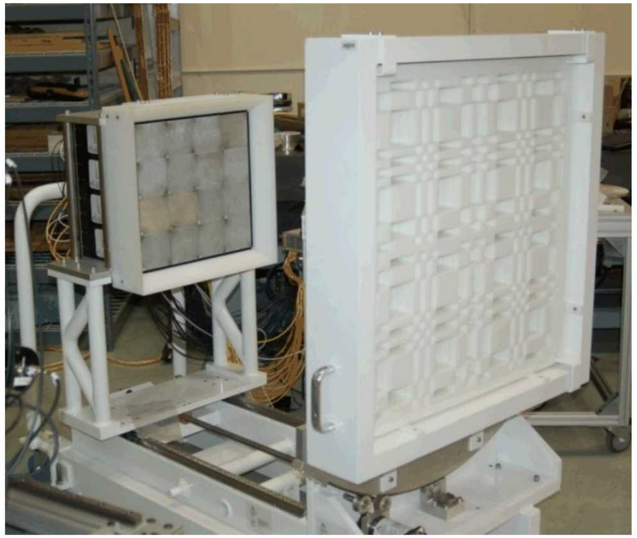
Radiation Detection at SNL

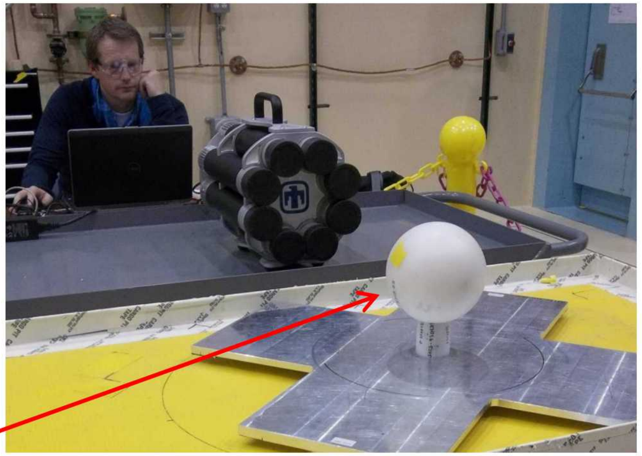
1D Time-Encoded Imager

- Large cells for high-efficiency
- Designed to localize weak sources at large standoffs

Beryllium Reflected Plutonium (BeRP) ball in a 1" shell of High Density Polyethylene

Neutron Coded Aperture Imager





8-Shooter Stilbene Array

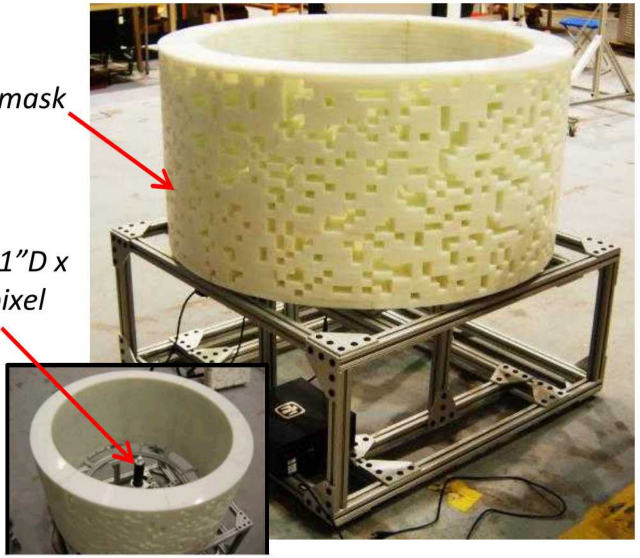
- Correlated n/γ analysis based on fast timing

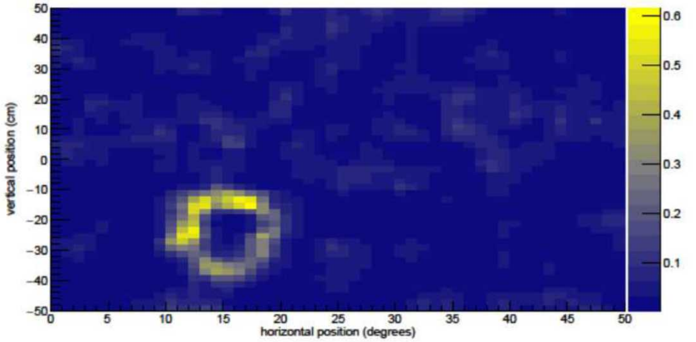
- Imagers based on encoded masks increase the total image resolution
- Reduced per-event resolution and energy resolution

2D Time-Encoded Imager

2D coded mask

Single 1"D x 1" LS pixel



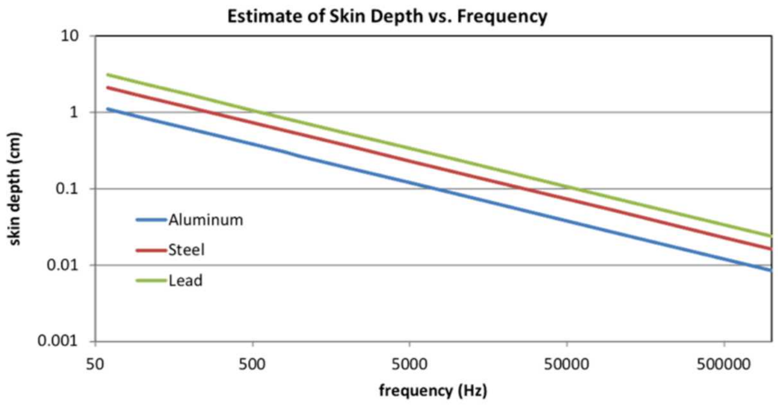


^{252}Cf source moved through an extended pattern at 2-m standoff

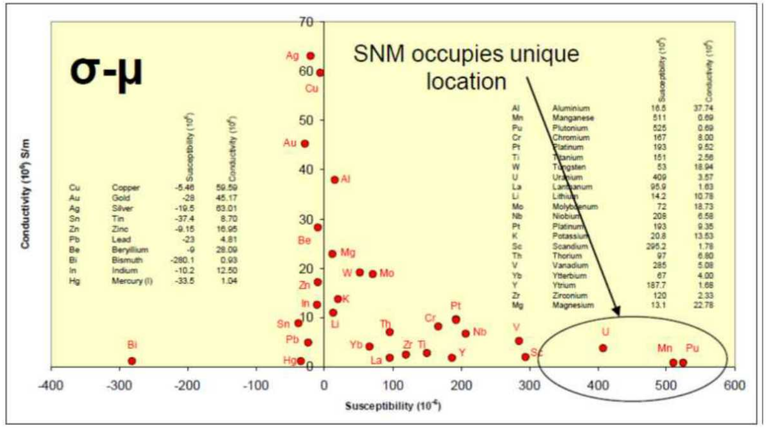
Alternative Signature Detection

Electromagnetic Signatures

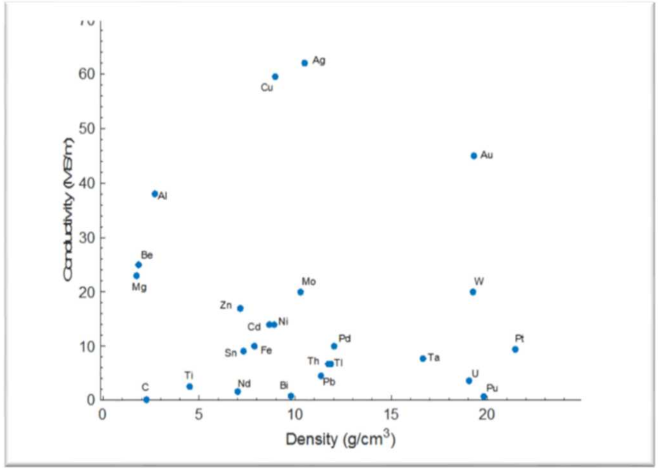
- Electromagnetic properties provide a complementary signature to traditional radiation detection techniques for detecting and characterizing special nuclear material and other threats
- Potential applications spaces include:
  - Container screening
  - Detection of shielded SNM
  - Detection of hidden objects/voids
- Low-frequency AC magnetic fields can be used to penetrate through shielding materials
- Magnetic induction tomography (MIT)probes the conductivity of the material
- Susceptibility and nuclear magnetic resonance (NMR) signatures are a potential path towards improved specificity



Approximate skin depth as a function of frequency for common shielding materials



Conductivity vs. susceptibility for select materials

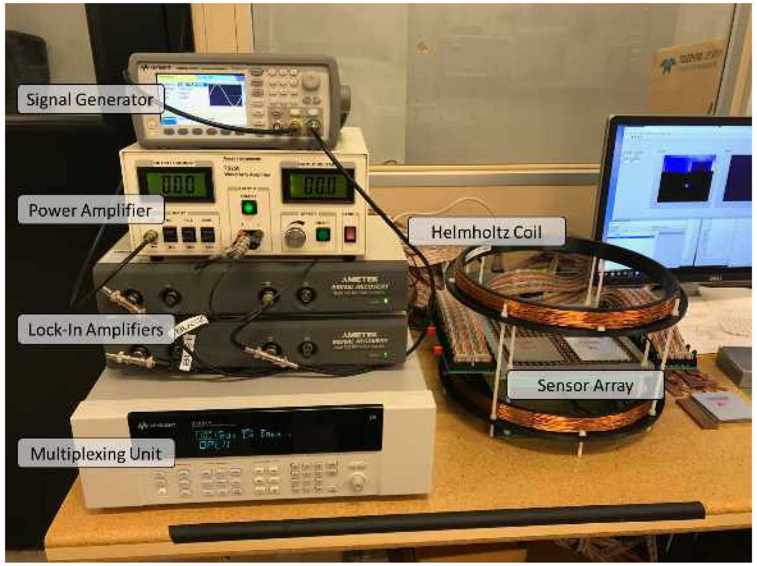


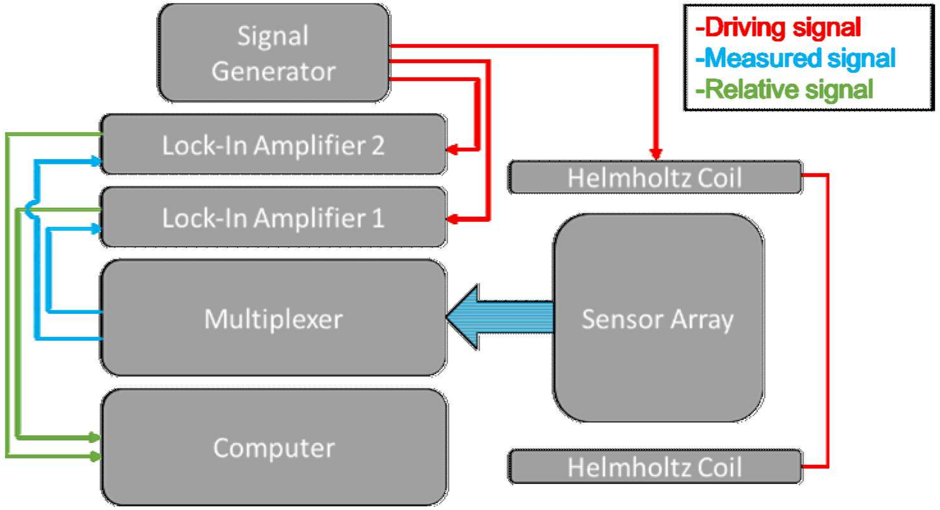
Conductivity vs. density for select materials

Magnetic Induction Tomography

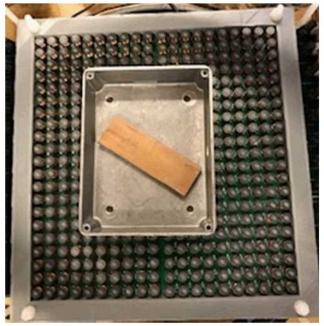
Imaging Array Concept

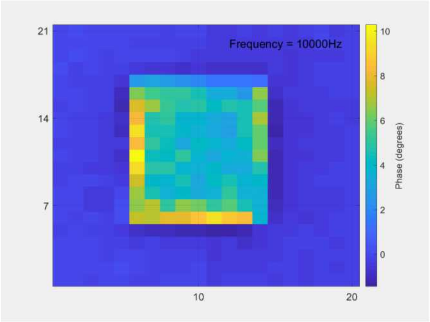
- Helmholtz coil with AC driving signal generates uniform field across sensor array plane
- Induced eddy currents in conductive objects perturb the field
- Lock-in amplifiers compare measured signal relative to driving signal
- Multiplexer steps through each sensor coil sequentially to build 2D pixelated image





Low-frequency penetration of conductive shielding

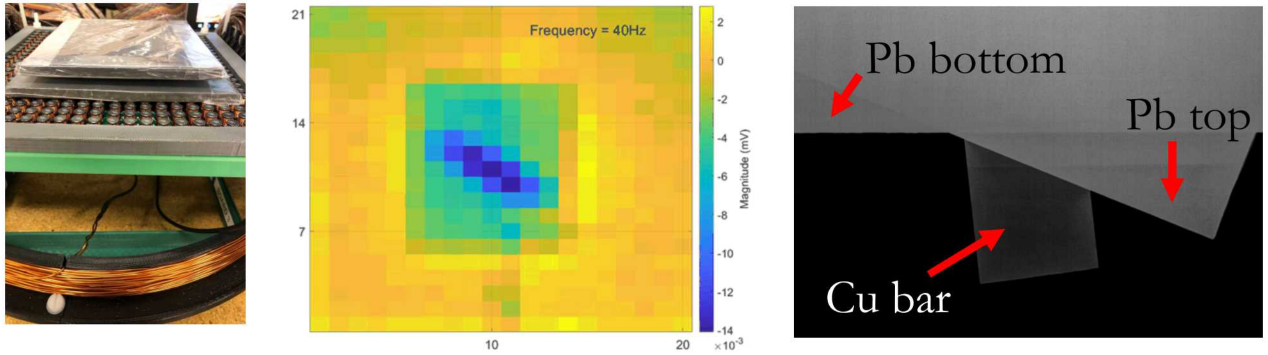




Magnetic Induction Tomography

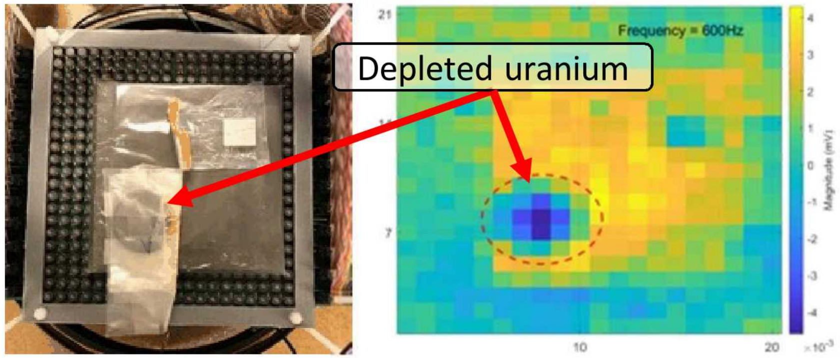
Imaging Array Results

Magnetic Induction Tomography vs. x-ray Radiography



A 0.2” Cu bar is visible between two 0.25” thick lead sheets in a 40 Hz MIT image but is not visible in a 3-minute 450 kVp x-ray image

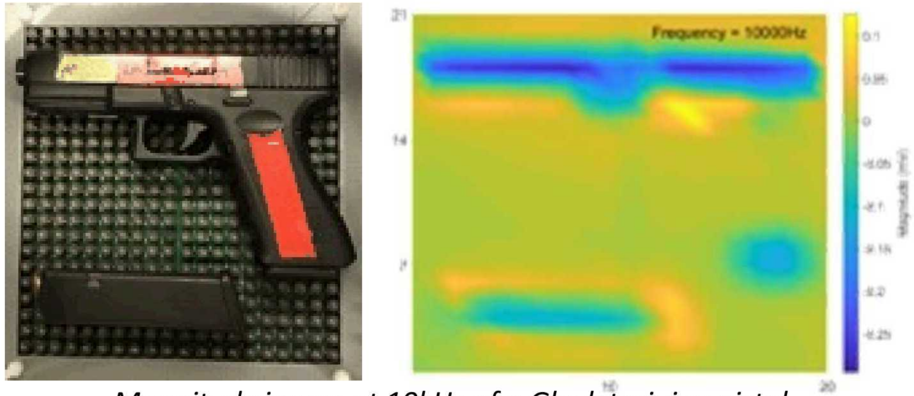
Depleted Uranium Shielded by 0.25” Lead



Detection of lead-shielded depleted uranium through 0.5 cm thick lead sheet using implicit background subtraction

(σ_{Pb} = 4.6 MS/m, σ_U = 3.6 MS/m)

Firearm Screening

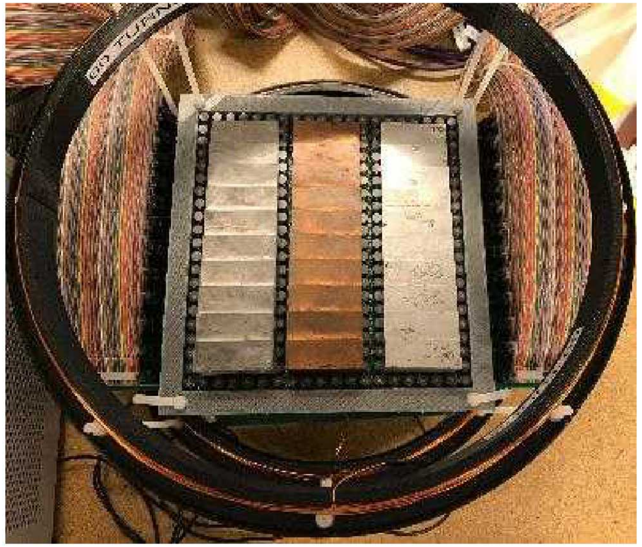


Magnitude image at 10kHz of a Glock training pistol

Step-Wedge Measurements
Material Characterization

- Various radiography step-wedge samples with thicknesses between 0.1” and 1”
- Samples imaged in threes with copper in the middle for reference
- Frequency swept between 100 and 10k Hz in 27 logarithmic steps

Radiography sample collection



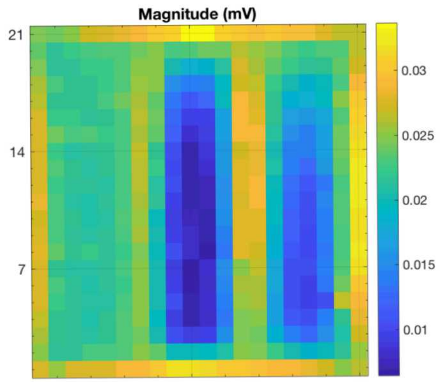
Lead-Copper-Aluminum

Measurement sets

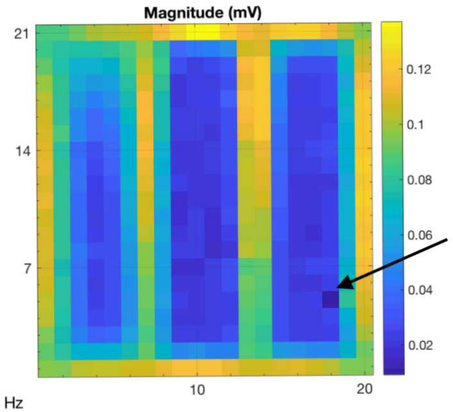
Group #	Sample 1	Sample 2	Sample 3
1	Lead	Copper	Aluminum 6061
2	Stainless Steel	Copper	Titanium
3	Wood	Copper	Carbon
4	Beryllium	Copper	Magnesium
5	Plexiglas	Copper	Epoxy
6	Tungsten	Copper	Tantalum

Step-Wedge Measurements
Lead-Copper-Aluminum

Image progression as a function of frequency

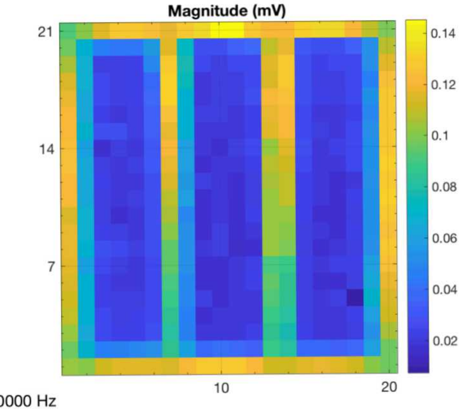


100 Hz



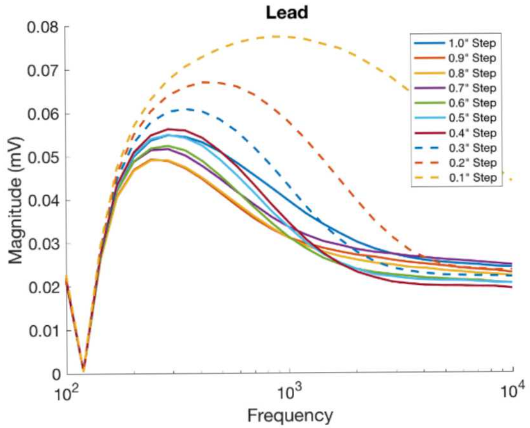
1000 Hz

Piece of chipped ferrite core

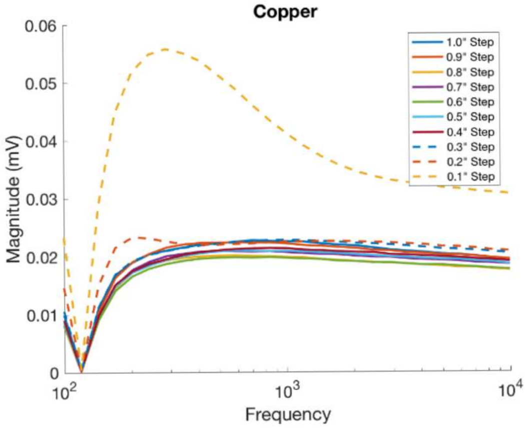


10000 Hz

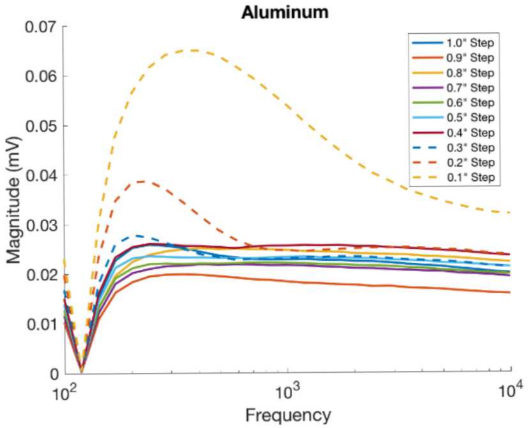
Frequency-dependent response at each step thickness



σ_{Pb} = 4.6 MS/m



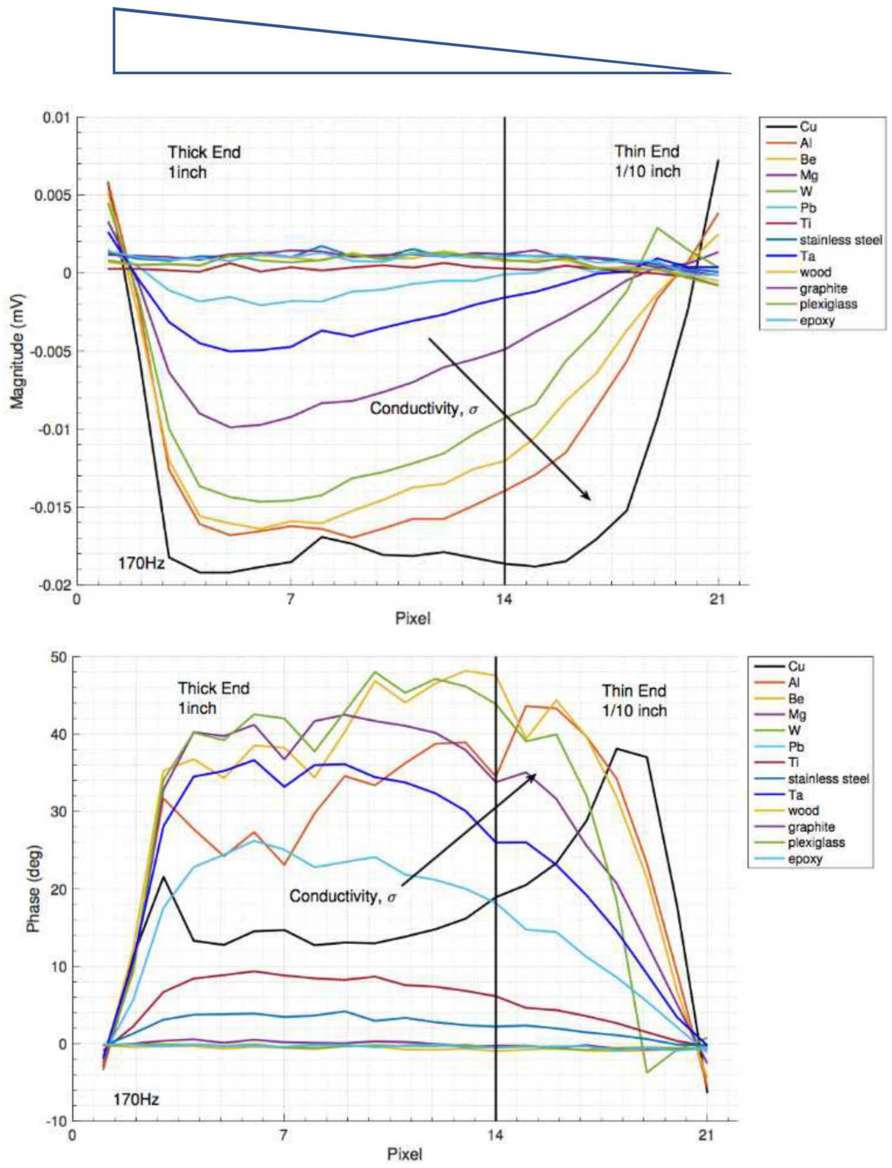
σ_{Cu} = 60 MS/m



σ_{Al} = 38 MS/m

Step-Wedge Measurements

Dependence on material thickness



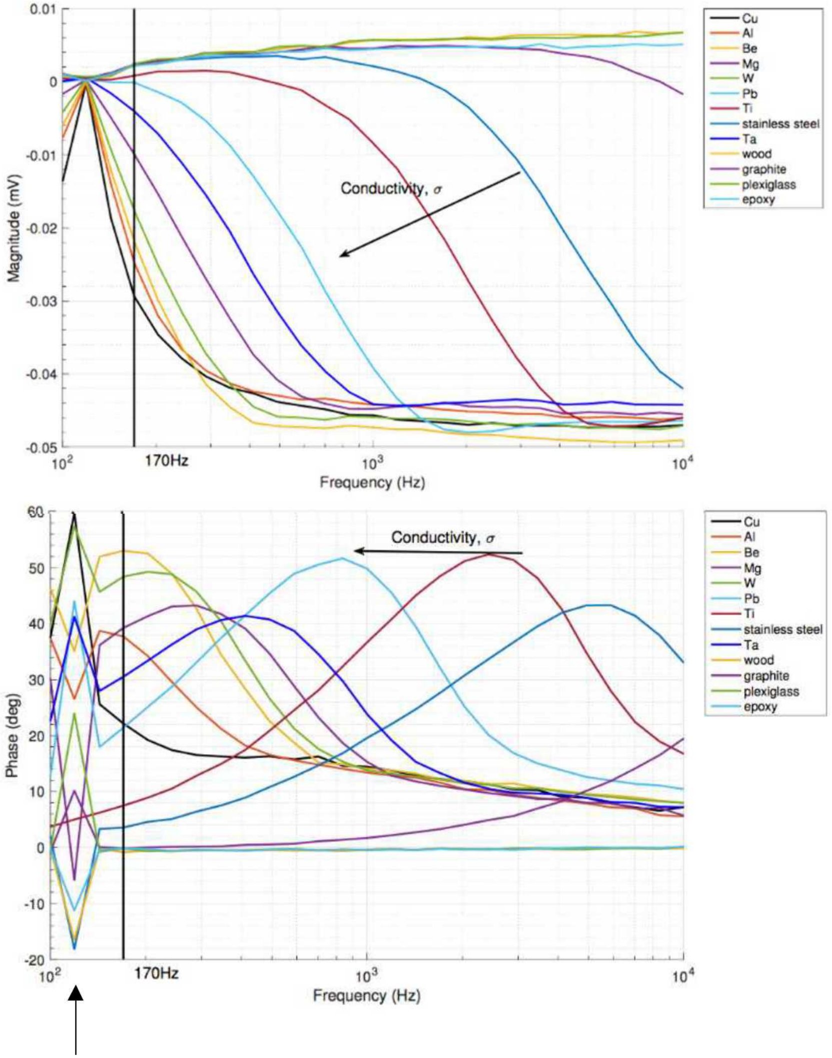
- Line trace for each material shows the penetration of the eddy currents as a function of thickness
- Skin depth is inversely related to conductivity and frequency

$$\delta = \sqrt{2/\omega\sigma\mu}$$

- Phase shift increases with material conductivity
- All data is background subtracted and can be calibrated to copper control

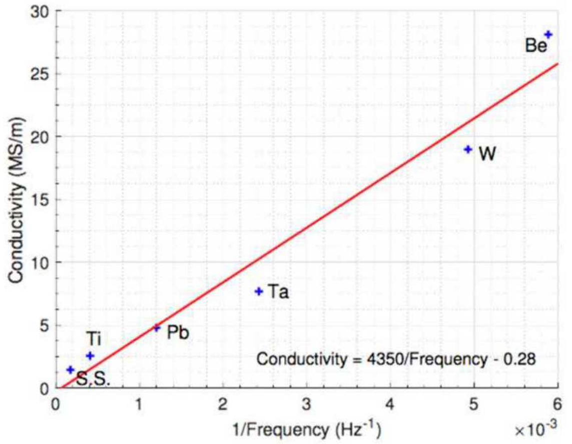
Step-Wedge Measurements

Dependence on driving frequency



120Hz mains noise

- Frequency-dependent response shown for the 0.4” step
- Signal magnitude falls off faster for materials with higher conductivity
- Phase shift shows resonant frequency that varies between materials
- Analysis demonstrates correlation between resonant frequency and conductivity



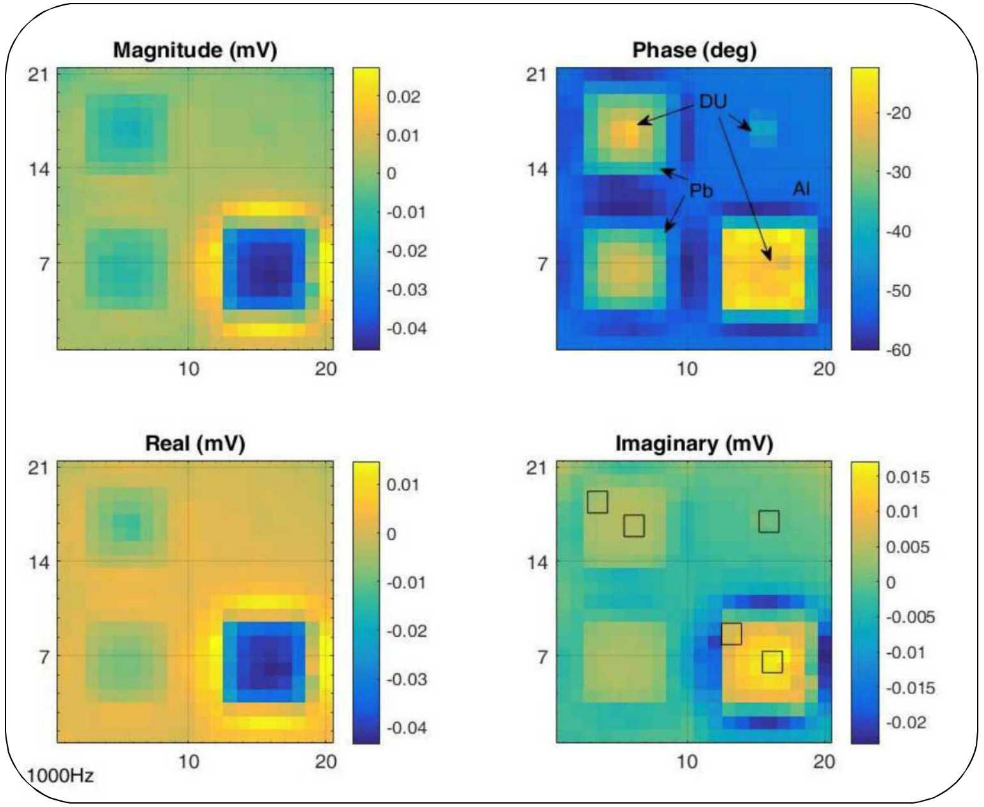
Measurements of Depleted Uranium

Penetration through shielding

Measurement of smaller DU samples allowed for simultaneous analysis of shielding and DU

- Unshielded DU coupon
- Lead-shielded DU coupon
- Aluminum-shielded DU coupon
- Lead-shielded DU bullet

Various DU samples and shielding coupons

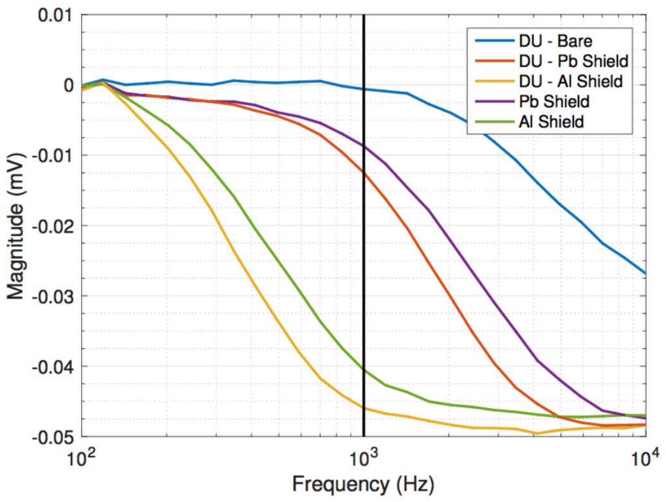


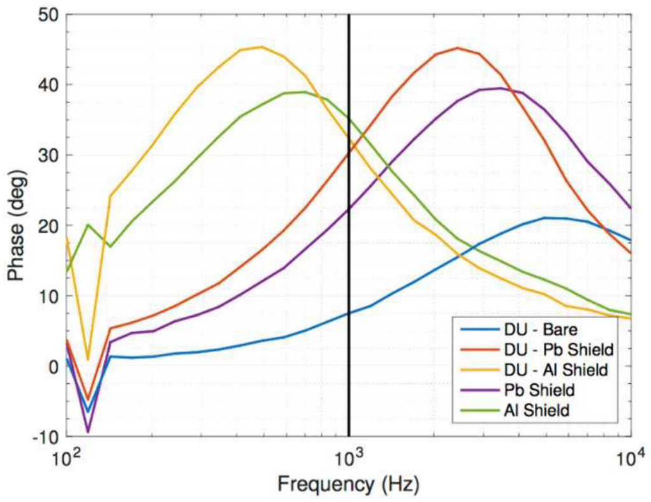
Raw data collected at 1000 Hz
Boxes in imaginary image denote individual pixels analyzed

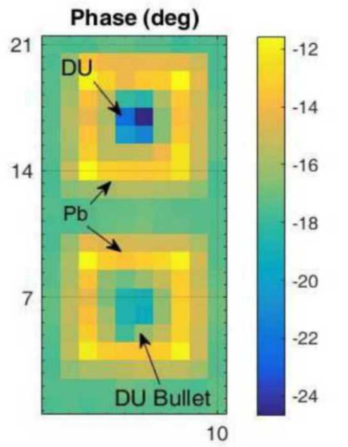
Measurements of Depleted Uranium

Dependence on driving frequency

- Frequency-dependent response investigated at five pixels
- Induced eddy currents appear to undergo a resonance, resulting in a peak in the relative phase measurement
- Resonant frequency can be used to inform dual-frequency correction and suppress shielding in the image







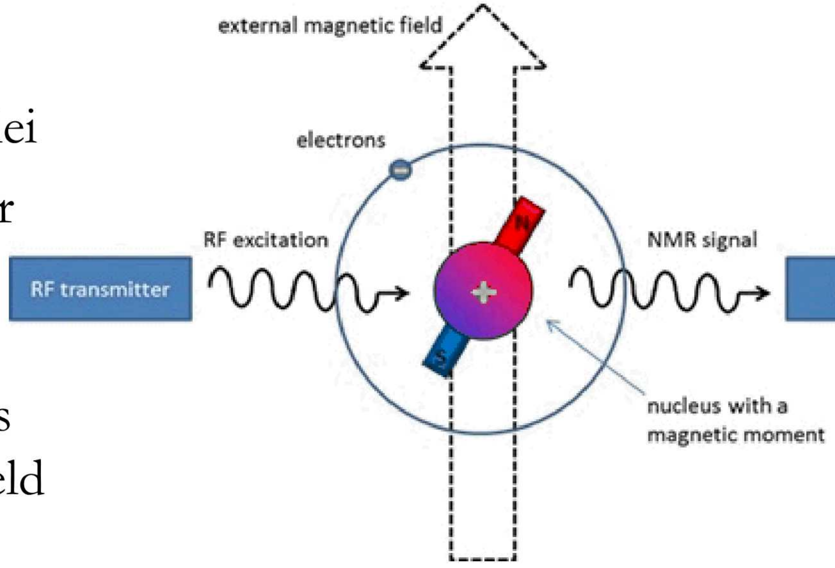
Dual-frequency corrected (2.42 - 3.46) kHz

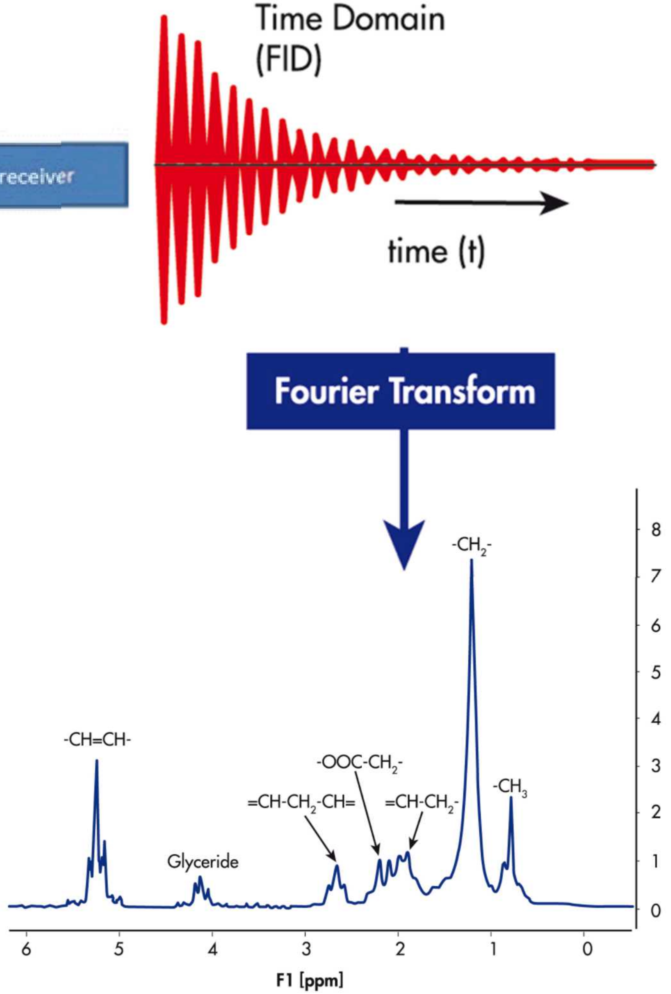
Alternative Signature Detection

Nuclear Magnetic Resonance Spectroscopy

NMR Detection Technique

1. Apply a magnetic field to align the nuclei
2. Apply an excitation pulse at the Larmor frequency to tip the atom into the transverse plane
3. Measure the magnetic field produced as the nuclei precess about the aligning field
4. The frequency spectrum of the measured signal is used to characterize the sample



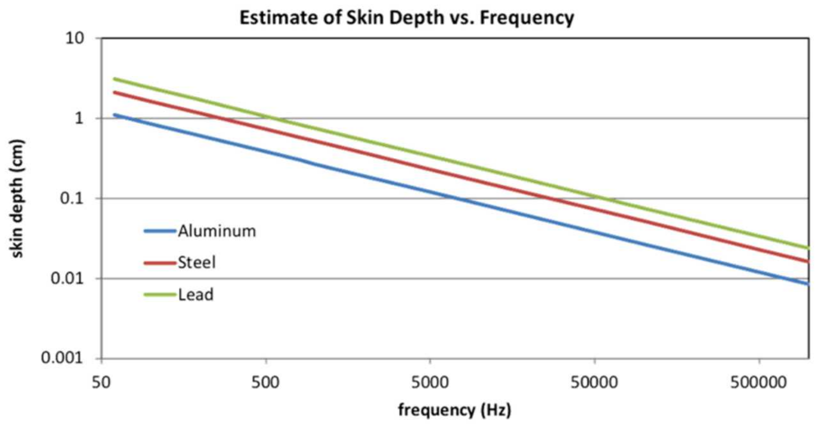


http://www.scientistlive.com/content/benchtop-nmr-spectroscopy

- The spin quantum number depends on A and Z of the nucleus (e.g. ^1H has spin = ½ and gyromagnetic ratio of 42.6 MHz/T)
- Typical high-resolution NMR measurements are performed using multi-Tesla aligning fields with Larmor frequencies in the MHz range (e.g. in a 7T field ^1H has a Larmor frequency of 300 MHz)

Low-Field NMR
Motivation

- NMR is a standard tool for identifying the chemical nature of organic compounds, but can be applied to any nuclei with and odd number of protons and/or neutrons
- Earth's field NMR utilizes low-frequency ($\lesssim$ 2 kHz) excitation pulses to stimulate nuclei at their characteristic Larmor frequency
- Coil array work has demonstrated how low-frequency magnetic fields can be used to penetrate shielding
- Can we apply this concept to nuclear magnetic resonance and use low frequency EM interrogation to detect and potentially characterize shielded threat objects?



Approximate skin depth as a function of frequency for common shielding materials

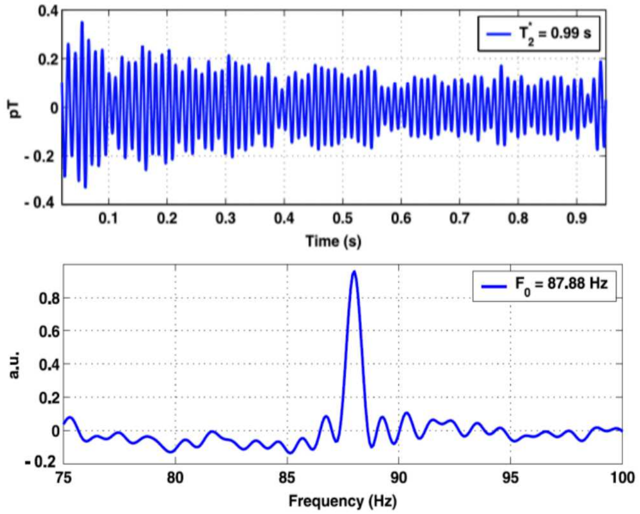
	Isotope	Natural abundance (%)	Gyromagnetic ratio (MHz/T)	Larmor frequency at Earth's field (Hz)
Select Explosives Isotopes	^{1}H	99.98	42.6	2059.2
	^{13}C	1.1	10.7	517.9
	^{14}N	99.6	3.02	148.9
	^{19}F	100	40.1	1937.1
	^{31}P	100	17.2	833.6
	^{35}Cl	76	4.13	202.0
SNM	^{235}U		-0.79	40.0
	^{239}Pu		3.02	149.3

NMR-relevant properties for common isotopes commonly found in explosives and SNM

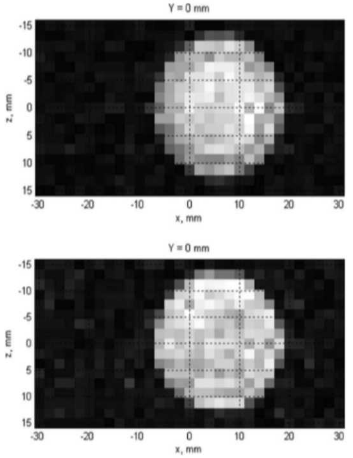
Low-Field NMR

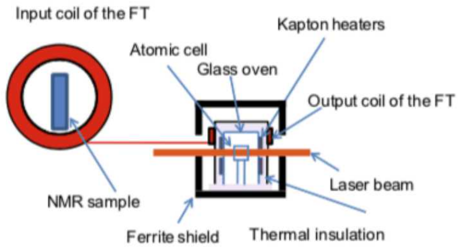
Motivation

- The signal strength is expected to be in the pT-fT range, which will require high-sensitivity magnetometers for detection
- A large body of work exists using SQUID magnetometers, including the detection of the ^{1}H NMR signature through 2 mm of copper
- SQUIDS require cryogenic cooling, making them difficult to field
- Commercial availability of atomic magnetometers present an opportunity to develop a field-deployable system (if we can get high enough sensitivity at earth's field)



Water in a 2mm thick copper canister, 2 μT B_0, read out by a SQUID. (Matlachov et al. 2004. LANL)





MRI at 3.2 kHz using a SQUID (top) and an atomic magnetometer (bottom). (Savukov et al. 2009. LANL)

Challenges

- Earth's field is uniform enabling larger "samples", but the NMR signatures will be weakened and broadened due to low-aligning fields, shielding, standoff, and the potential for solid state materials
  - High sensitivity magnetometers are a must
- Shielded materials may be subject to an an unknown alignment field
  - NMR signatures might be identified by sweeping through a range of frequencies
- Chemical shifts are typically used to determine the molecular form of a sample, but these are not detectable at low fields,
  - J-couplings are available at low fields, and can help with characterization (if we can detect them)
- Commercially-available vector atomic-magnetometers do not yet operate at Earths field and do not have an large enough sensitivity bandwidth
  - Might be able to perform active field cancellation
  - RF-tuned magnetometers may help

Low-Field NMR

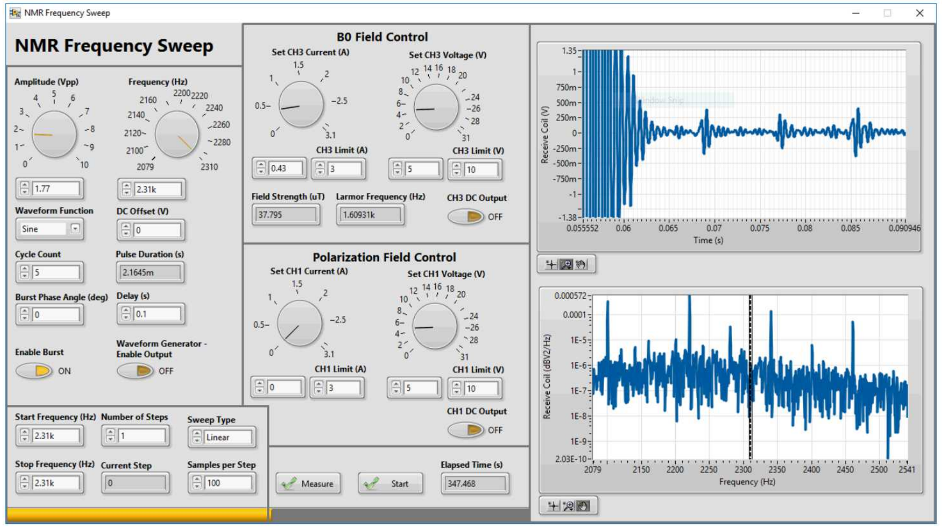
Project Objectives

1. Standup low-field NMR tested
   - Demonstrate detection at controlled-fields at and below Earth's field using pickup coils
   - Replace receive coil with QuSpin Gen-2.0 QZFM for improved sensitivity and drop fields to $\leq$ 200nT
2. Explore options for improving functional range of high-sensitivity detection at Earth's field
   - Expanded bandwidth and external field cancelling coils to increase operating background
   - Move toward RF-tunable magnetometer
3. Demonstrate detection of ^{1}H NMR signature of shielded samples at Earth's field
4. Demonstrate detection of other shielded isotopes in solution state at Earth's field
5. Demonstrate detection of chemical signatures using J-coupling to help identify chemical form of sample
6. Demonstrate detection of NMR signatures in solid-state
7. Perform measurements on kilogram quantities of SNM and other relevant threat objects

Low-Field NMR Progress

Development of a *Low-field NMR Testbed*

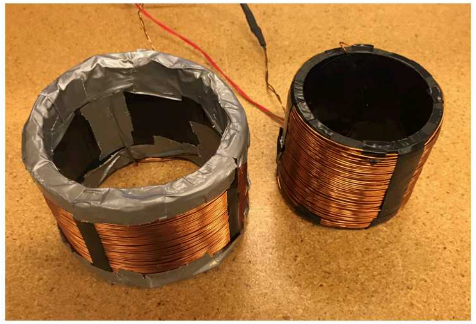
- Initial design is based on the setup described in: C. A. Michal, "A low-cost spectrometer for NMR measurements in the earth's magnetic field," (2010) Data acquisition software being developed in LabVIEW
- Three separate coils used for pre-polarization, transmit, and receive
- LabVIEW interface allows control of transmit and pre-polarization coils
- Transmit signal generated by an Agilent 33250A function generator operated in burst mode and passed through a current divider
- Receive signal (coil or SERF) read out using 14-bit 100 kS/s NI-9215 ADC
- Voltage induced on receive coil is expected to be in the 100s of nV range, so is passed through an amplification/bandpass circuit with a gain of 50000



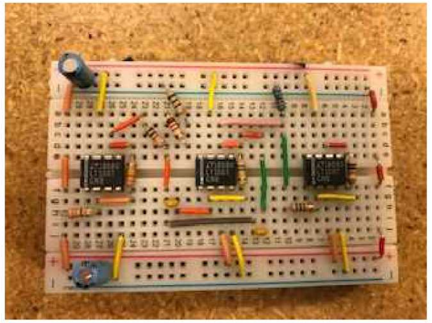
Custom LabVIEW Interface

Coil	Radius (cm)	Height (cm)	Turns	Wire Gauge	Calculated Inductance (μH)
Transmit	4.5	7.2	46	18	148
Receive	4.5	7.2	1100	30	84851
Pre-polarization	5.7	7.2	180	18	3389

Coil properties



Pre-polarization, transmit and receive coils



Receive coil amplification and bandpass circuit

Low-Field NMR Progress

Atomic Magnetometer Hardware

- QuSpin Zero-Field Magnetometer – Gen 2.0
  - Vector magnetometer
  - 7-10 fT/√Hz sensitivity
  - 135 Hz bandwidth
  - 200 nT max background

QuSpin Gen-2.0 QZFM

Ambient field measurement by QZFM inside shielding

TwinLeaf MS-1L Magnetic Shield

- Four-layer MuMetal Shield
- Shielding factor: 10^6
- Magnetic noise: 16: fT/√Hz
- Internal field coils
  - 3-axis magnetic field coil to provide fields up to ~50 μT (i.e. Earth's field)
  - Also has 5-axis gradient field coil and Z-axis second order gradient coil

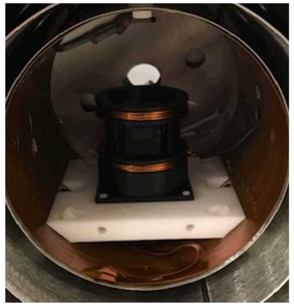
Twinleaf MS-1L shield with QuSpin Gen-2.0 QZFM inside

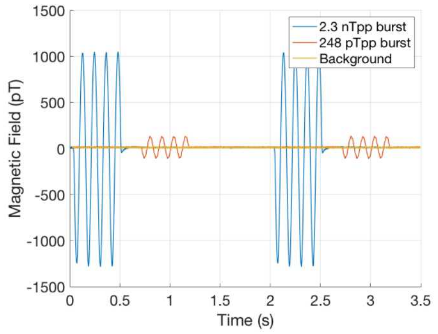
Low-Field NMR Progress

Atomic Magnetometer Testing

Low-Field Pulse Testing

- Larmor frequency of ^1H at 200 nT is 8.5 Hz
- A 5 turn Helmholtz coil driven by a Keysight 33600A waveform generator was used to provide a low-amplitude, 8.5 Hz magnetic field pulses in the detectable range of the QZFM

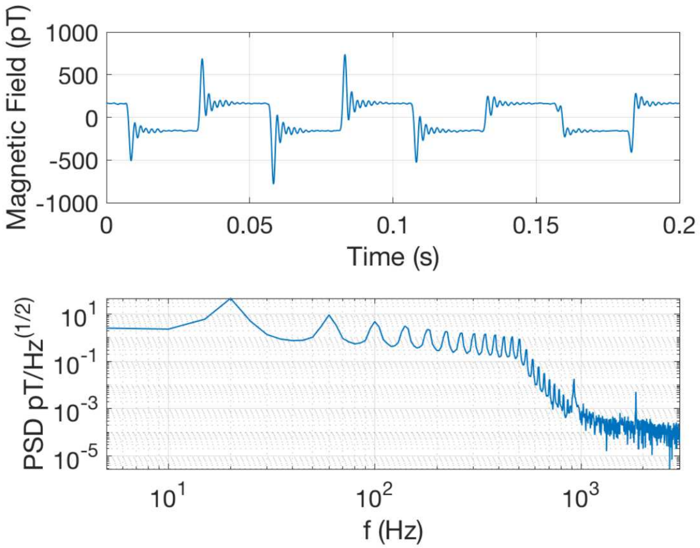




Low-amplitude 8.5 Hz pulses generated by 5-turn Helmholtz coil and detected by QZFM

QZFM Frequency Response

- A 20 Hz square wave was used to test probe the frequency response over a larger bandwidth
- Standard analog output is limited by a low-pass filter
- Savukov et al. [1] who demonstrated sensitivity up to 1.7 kHz by bypassing the low-pass filter and manually post-processing the photodiode output



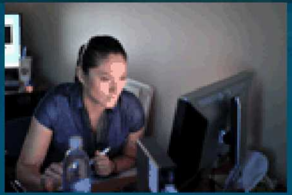
Response of QZFM to 20 Hz square wave generated by 5-turn Helmholtz coil

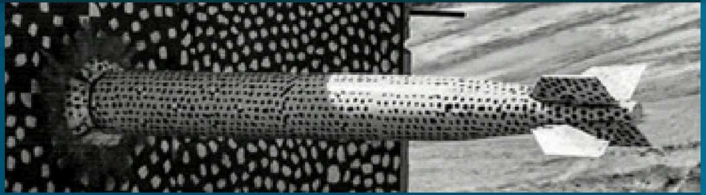
Summary

- High-risk, high-payout endeavor could ultimately lead to tools for the detection and characterization of high-explosives and shielded SNM in sealed containers
- Mission success would result in a new capability would meet the needs of multiple sponsor mission areas including
    - Emergency-response diagnostics
    - SNM search/screening
    - Arms control
    - Safeguards
- This is a challenging problem that requires both proof-of-concept in the applications space as well as continued development of a suitable atomic magnetometer
- Many of the challenges of this problem are being addressed individually by various research groups, but the combination is likely unique to our desired application space
- Combining mission knowledge with technical expertise through collaborations across SNL and with external partners will drive innovation and increase the likelihood of a successful outcome

Earth's-Field NMR for the Detection and Characterization of Shielded Threats

7 August 2019





PRESENTED BY

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