Imaging of Individual Barium Atoms in Solid Xenon

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Why do we need to detect individual Ba atoms in solid xenon?

We are interested in doing the most sensitive neutrinoless double beta decay experiment possible to discover this process. Example:

\[ ^{136}Xe \rightarrow ^{136}Ba^{++} + 2e^- \]

Normally you would also have two anti neutrinos emitted.

The discovery of neutrinoless double beta decay changes the fundamental understanding of elementary particles:

- Implies neutrinos and antineutrinos are the same particle
- Could be basis for “leptogenesis” in the Big Bang: why the universe is composed of matter only
- Information on neutrino masses
Extending the Sensitivity of Neutrinoless Double Beta Decay in the nEXO Detector

\[ ^{136}Xe \rightarrow ^{136}Ba^{++} + 2e^- \]

- Ba Tagging: also detect the daughter \(^{136}\)Ba ion or atom located at the decay position.
- Potential to eliminate all but 2νββ backgrounds

*Other backgrounds, e.g., γ ray, don’t produce a Ba atom.*

Current experiments detect only the emitted electrons and measure their energy accurately.
Current EXO-200 experiment: 200 kg of enriched LXe

51 total events detected in expected energy range in 3 years
~44 are due to background.
What if you could eliminate all background?
7 events with no background might constitute a discovery!

Energy of 2 electrons in 0νββ decay

0νββ decay half-life limit: $1.8 \times 10^{25}$ years
0νββ decay half-life sensitivity: $3.7 \times 10^{25}$ years

Next generation experiment nEXO: 5000 kg of enriched LXe: with Ba tagging expect $>10^{28}$ year half-life sensitivity
Ba tagging in solid Xe concept

- Detector locates the decay position.
- Capture Ba using a cryogenic probe to trap Ba in solid Xe.
- Extract probe and do laser spectroscopy on probe at lower temperature.
- **Count Ba atoms:**
  - 1 = $\beta\beta$ decay; 0 = not $\beta\beta$ decay
Ba$^+$ ion beam deposition apparatus

Induction plates detect pulses without blocking the beam

1. Cool sapphire window to 50K
2. Begin Xe gas flow for a few s
3. Pulse Ba$^+$ beam onto window
4. Stop Xe gas flow after a few s
5. Cool window to 10K

Source of Ba$^+$, Selects Ba$^+$, Focus, deflection, and deceleration, Pulse beam and detect at $\mu$sec level
Observation

- Excite with tunable dye laser
- Observe fluorescence thru filter
- Image with LN-cooled CCD
- Have ability to scan laser with piezo-electric translation stages
- Evaporate sample at 100K
Energy levels of Ba in vacuum

Possibility of bleaching due to optical pumping

- $6s6p \ ^1P_0$
- $6s6p \ ^3P_{0,1,2}$
- $791.1 \text{ nm}$
- $553.5 \text{ nm}$
- $6s^2 \ ^1S_0$
- $6s5d \ ^1D_2$
- $6s5d \ ^3D_{1,2,3}$

Matrix transitions and lifetimes:

- $1/300$
Spectra of Ba in SXe

Ba fluorescence spectra: 4 different matrix sites

- 3 Ba peaks experience bleaching after absorption of $10^4$-$10^6$ photons
- Ba atoms in the matrix site with 619-nm fluorescence have low bleaching

Ba excitation spectra of different matrix sites

- 2 Ba sites have characteristic 3-peak excitation spectrum.
- 572 nm

Progress in theoretical calculations that explain the excitation spectra of two of the Ba in sXe lines

Emission spectra have not yet be simulated.

Raw CCD image of a few Ba atoms
Small amount of background from sapphire window surface is primary competition to single Ba imaging

**Background Bleaching**

- 532 nm laser rastered across sample (90μm × 90μm) overnight at 100 K
- Reduces surface background by a factor of 30
Fixed Laser Images

- Fluorescence signal is linear with # of ions deposited: not Baₙ molecule!
- Best fit of 379±10 counts/mW/s/ion
Scanned Images of Ba Atoms

Each camera exposure is for a position in a grid.

See peaks as laser moves over single atoms
Scanned Images of Ba Atoms

Each camera exposure is for a position in a grid:

See peaks as laser moves over single atoms

Moving laser in x in 4 µm steps
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

(a) Laser rastered across sXe only deposit: no peaks. *Deposit evaporated.*

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

SXe Deposit evaporated.
b) Laser rastered across Ba/Xe deposit: strong fluorescence the laser positions of two individual Ba atoms.

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

c) Repeat scan: single atom peaks persist.

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

Counts in laser beam region per CCD frame

Then laser moved to the left peak. 3s images are taken for 150 s.
Atoms emit for ~25s more, then turn off: (>300 s for other atoms)

Lots of photons emitted by one Ba atom! (700,000 - >10^7)

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

d) Signal of one atom gone in repeat scan.

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Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

Deposit was $44^{+4}_{-11}$ Ba$^+$ ions in 48 μm x 48 μm scan area.

Reminder:
- Don’t know conversion efficiency Ba$^+$ -> Ba
- Some Ba are in the other 3 matrix sites

d) Signal of one atom gone in repeat scan.

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Composite scan image
(each pixel is integral of counts in 3x3 laser region of CCD image)

Deposit evaporated.
e) Scan of sXe only deposit has no peaks (σ of sXe deposits in 3s is ~15 counts).

**Ba deposit can be “erased” by evaporating and re-freezing the solid Xe coating.**

*No sensitivity to any stray Ba atoms on window surface.*

C. Chambers et al., arXiv 1806.10694, submitted to Nature
Even in a large Ba deposit of ~7000 Ba$^+$ ions, evaporation "erases" the Ba signal.
Time Resolved Photon Counting for background reduction

- Excite with 100ps pulsed 560nm diode laser
- Record arrival time of photons at SPAD
- Ba decay lifetime is $7.0 \pm 0.3$ ns
- Window surface background decay lifetime is $\sim 3.5$ ns
- SS Cryoprobe tube decay lifetime is $\sim 1.5$ ns

Some possible discrimination against window surface background. Good discrimination against cryoprobe metal tube background.
Comments on interpretation of the 619 nm emission peak as Ba atom:

We have looked at alternate hypotheses:
- Ba\(_n\): dimer, trimer, ...: linearity of signal vs. #ions deposited refutes this.
- Ba\(^+\): spectrum is same for Ba atom and Ba\(^+\) ion source
- Lattice damage: no peak for similar Ar\(^+\) deposit
Comments on interpretation of the 619 nm emission peak as Ba atom:

We have looked at alternate hypotheses:

- Doesn’t match transitions in Ba molecules:
  - BaH$_2$\(^+\)? (sharp, observed when deposit at 10 K but not in 50 K deposit)
  - BaO broad (blue-red), characteristic vibrational pattern in sNe, sAr missing
    - BaO lifetime 356 ns (\(A^1\Sigma\)) or 9 µs (\(a^3\Sigma\)) vs. 7 ns observed
  - BaOH (blue-green, IR emission in vacuum)
  - Deposit at 50 K should also reduce \(N_2\)\(^{15}\) \(O_2\) content in SXe

![Graphs showing emission and excitation counts with wave numbers](image1)


![Graph showing BaO chemiluminescence in Ar clusters](image2)

Interpretation of 619 nm peak matrix site

Ba atoms are too big to fit in single vacancy (SV) site.

Ba atoms known to exist in 4-vacancy (TV) and 5-vacancy (on grain boundary) sites.

Favored interpretation from the chemists (John McCaffrey personal communication): Ba\(^+\) forms a tighter bond with Xe and is energetically favored to be in a single vacancy (SV) site. Hypothesis: Ba\(^+\) neutralizes to Ba after it forms in the SV site. Then it is energetically unfavored to move additional Xe out of the way.
Apparatus to capture Ba in Sx on a cryoprobe

Probe insertion/retraction mechanism:
- Large motor-driven bellows
  - Removes probe from LXe
- Spectroscopy at lower temperature
  - Gate valve
  - Observation Volume with pumpout valve

After close gate valve, increase probe cooling and reduce Xe gas pressure, following gas-solid vapor pressure curve -> 10K.

Successful freezing from LXe and extraction to observation volume
Implantation of $\text{Ba}^+$ ions into S$\text{Xe}$ on cryoprobe

- $\text{Ba}^+$ ions created by laser ablation in Xe gas
- E-Fields to drift ions into L$\text{Xe}$ and to Cryoprobe.

- Spectroscopy of Ba in S$\text{Xe}$ in observation chamber still to be done
Conclusions

• Individual Ba atoms have been imaged in solid noble gas matrix
• Possibility to image Ba atoms in other matrix sites with $10^4$’s to $10^5$’s photons
• Don’t see Ba atoms on the window surface or left over from a previous deposit.