Imaging small numbers of Ba atoms in solid xenon for barium tagging in nEXO

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Images of Ba atoms in solid Xe in a focused laser region, after deposition from vacuum onto a cold sapphire window, are obtained using a 619-nm fluorescence peak down to the single-atom level. This is an important step toward barium tagging with a cryogenic probe from liquid Xe for the nEXO neutrinoless double beta decay experiment.

I. INTRODUCTION

The search for neutrinoless double beta decay ($0\nu\beta\beta$) is an important probe into the nature of neutrinos. Observation would require that neutrinos are Majorana particles, would demonstrate violation of lepton number conservation, and could help determine the absolute neutrino mass [1]. EXO-200 is searching for $0\nu\beta\beta$ in ^{136}Xe with around 170 kg of liquid Xe (lXe) enriched to 80.6% ^{136}Xe in a dual time projection chamber (TPC). Two-neutrino double beta decay ($2\nu\beta\beta$) of ^{136}Xe has been observed by EXO-200, and its half-life is measured at $T_{1/2}^{2\nu\beta\beta} = 2.165 \pm 0.016(\text{stat}) \pm 0.059(\text{sys}) \times 10^{21}$ yr [2].

The most recent EXO-200 $0\nu\beta\beta$ search sets a limit on the half-life at $T_{1/2}^{0\nu\beta\beta} < 1.1 \times 10^{25}$ yr (90% CL), which corresponds to an effective Majorana neutrino mass of $\langle m_{\nu_e} \rangle <$190-450 meV, depending on nuclear matrix element calculations [3].

A lXe TPC has a unique opportunity to tag the daughter at the site of a double beta decay event, which would improve $0\nu\beta\beta$ sensitivity by effectively eliminating all backgrounds [4]. Ba tagging is being investigated for possible incorporation in the next-generation ton-scale lXe experiment, nEXO. A few techniques for Ba tagging are under investigation [5][hot probe paper?], including one for a possible future Xe gas TPC [6]. The method described here is for barium tagging in the nEXO lXe TPC, in which a cryogenic probe would be moved to the position of the $0\nu\beta\beta$ candidate in order to freeze the daughter ion into a small amount of solid Xe (sXe) at the end of the probe. It would then be detected by its laser-induced fluorescence in the sXe.

The spectroscopy of Ba in sXe is described in the previous work [7]. An image of $\leq 10^4$ Ba atoms was obtained with the strong fluorescence peaks at 577 and 591 nm. However, bleaching of these fluorescence peaks with laser exposure reduces the emission at these wavelengths rapidly at high laser intensity, e.g., using a focused beam. Thus obtaining large numbers of photons from single Ba atoms is difficult without a method to overcome bleaching, e.g., with repumping lasers. Imaging was not attempted in [7] with the peak at 619 nm, though this peak has weaker bleaching by many orders of magnitude. This peak is promising for more sensitive imaging of smaller numbers of atoms because larger numbers of photons per atom may be obtained.

It is expected that a Ba^{++} ion will neutralize once to Ba^+ in lXe, as the lXe conduction band gap is slightly less than the ionization potential for Ba^+ [4]. It is also not known whether the Ba^{++} might neutralize completely in the charge cloud following a $\beta\beta$ event. A new study of neutralization of alpha decay daughters in the EXO-200 lXe TPC is in the process of publication [ref? arxiv? example of arxiv ref in 2014 nu review, [185]]. As a result, the feasibility of detecting single Ba ions as well as atoms is still of interest. This work focuses on imaging single Ba atoms in sXe via the low-bleaching 619-nm fluorescence. Varying numbers of atoms are observed in a focused laser region down to the single atom level, *and images of separate single atoms on a single deposit are obtained by scanning the focused laser.*
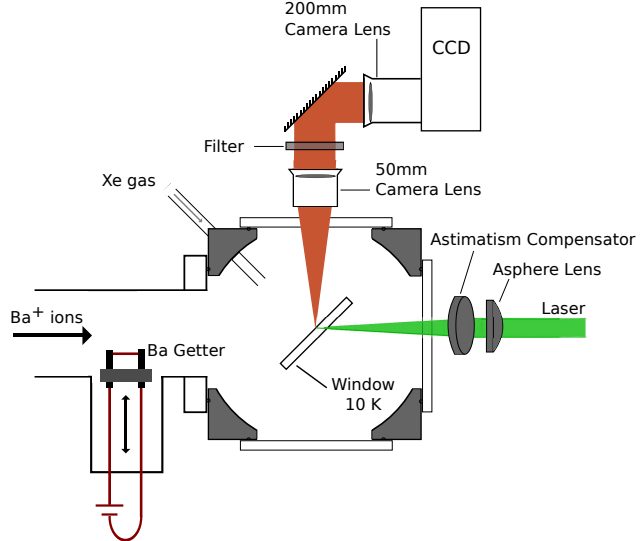

FIG. 1. Experimental setup for depositing Ba/Ba^+ in sXe matrices, and for excitation and observation. *want to put compensator in, and depict laser lens as asphere.*

II. APPARATUS AND METHOD

The apparatus for depositing and observing Ba/Ba^+ deposits in sXe is described in [7]. Important components are shown in Fig. 1. The source of Ba^+ is an ion beam at 2000 eV, filtered to select Ba^+ with an E×B velocity filter. A set of pulsing plates produces 1-μs pulses for depositing small numbers of ions. The spectra of Ba^+ ion deposits in the sXe matrix exhibit peaks known to be due to neutral Ba atoms [7]. Thus some percentage of the ions neutralize in the matrix, but the fraction has not yet been determined. An alternative source of neutral Ba is a $BaAl_4$ getter wire which can be inserted to emit Ba atoms toward the sample. However, it is challenging to achieve low Ba flux with this source and to calibrate it.

Deposits are made on a cold sapphire window tilted at 45°. To create a sample, Xe gas is directed toward the window using a leak valve. The Xe gas freezes on the window and forms a sXe matrix with a thickness of a few hundred nanometers. This is initiated a few seconds prior to the Ba deposit, continues during the Ba deposit, and is turned off a few seconds after the Ba deposit.

In this work, deposits are made with the sapphire window at around 50 K, partly to reduce hydrogen content in the matrix, as hydrogen freezes well below 50 K in vacuum [7]. The window is then cooled to 11 K for observation. Deposits made at 50 K result

in higher 619-nm signal than those made at 11 K, likely due to a higher population of 619-nm matrix sites. Xe deposition at around 30 nm/s also results in higher 619-nm signal, as compared to lower leak rates, while not being so high as to result in a frosty Xe matrix. An experiment cycle consists of a deposit at 50 K and a fluorescence observation at 11 K; then the deposit is evaporated by heating the window to 100 K. Many deposits are made in a day with varying numbers of ions deposited, as well as periodic Xe-only deposits to establish the background.

The excitation laser, a Coherent 599 cw dye laser with Rhodamine 6G dye pumped by the 514-nm line of a Lexel 3500 argon ion laser, enters from the back side of the window. Ba fluorescence light is collected and collimated by a 50 mm Nikon camera lens. A band-pass filter with FWHM of 20 nm passes just the 619-nm fluorescence peak. A 200 mm Nikon camera lens then focuses the image onto a liquid nitrogen cooled CCD, resulting in an image of 4x magnification. Each of the 20×20 μm pixels of the CCD represents approximately 5×5 μm on the SXe sample.

For a given laser intensity, the smallest focus possible is desired for optimal signal-to-noise from single atoms. To achieve this, an aspherical lens of 7.9 cm focal length is used to minimize spherical aberration, and a fused silica optical flat of 1 cm thickness is placed at $10°$ after the lens in order to compensate for astigmatism caused by the tilted sapphire window (see Fig. 1).

III.   RESULTS

A typical image made with a focused laser beam at 570 nm is shown in Fig. 2. The laser's path through the sapphire window is seen as a weak vertical line. At both ends, some background emission from the surfaces is observed. At the front surface (at the top in the image), extra fluorescence is detected from a small number of Ba atoms within the focused laser.

The number of Ba^+ ions deposited within the 1/e radius of the laser beam gives a rough upper limit to the number of Ba atoms responsible for the observed signal. For ion pulses of 13 fC/pulse and a focused laser $1/e^2$ radius of $w_0 = 2.1$ μm *I get 2.06 μm for x and 2.66 μm for y, 2.36 μm average*, this results in about 0.05 Ba^+ ions/pulse in the above area. The signal in Fig. 2 corresponds to a deposit of about 14 Ba^+ ions into the laser region, and is therefore roughly due to ≤ 14 Ba atoms. A ≤ 56-atom deposit (56 ions deposited) is shown in Fig. 3b, with Xe-only deposits made before and after (Fig. 3a,c)). It
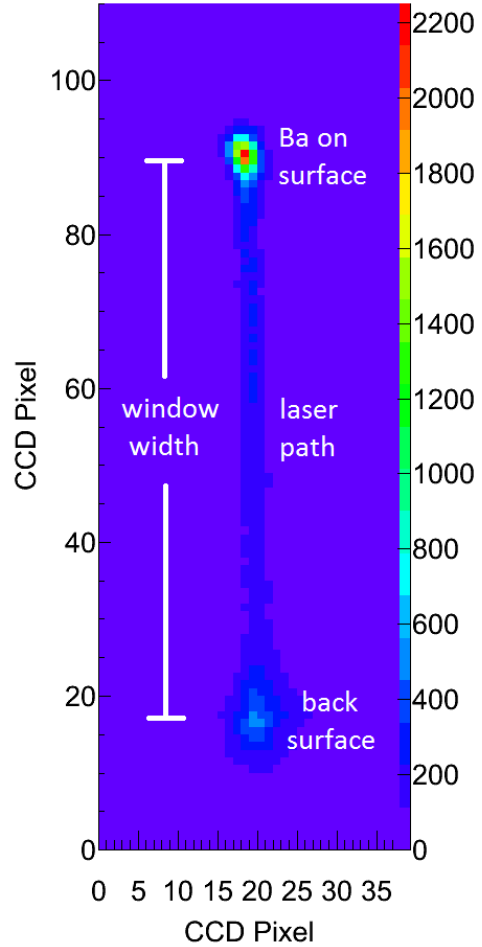

FIG. 2. Example image of Ba^+ deposit in focused laser. Pixels are 5 μm$\times$5 μm.

is important that no Ba is left over after evaporating the sample, even for larger deposits.

The counts vs. ions deposited are plotted in Fig. 4. Observe the linearity in summed CCD counts, from the image of the laser region in the sXe, vs. ions deposited for several deposits. Each point has subtracted the counts from a nearby Xe-only run. The observed signal corresponds to about 2000 counts/mW per atom with 60-s CCD exposures. Counts are scaled by laser power to account for small variations in laser power.

To obtain images of Ba fluorescence, sXe-only images are subtracted [This isn't said in explanation of linearity plot because those do not directly subtract images, they subtract integrated counts after using integration regions specific to each run (due to slight movements)]. A subtracted image of a deposit cor-
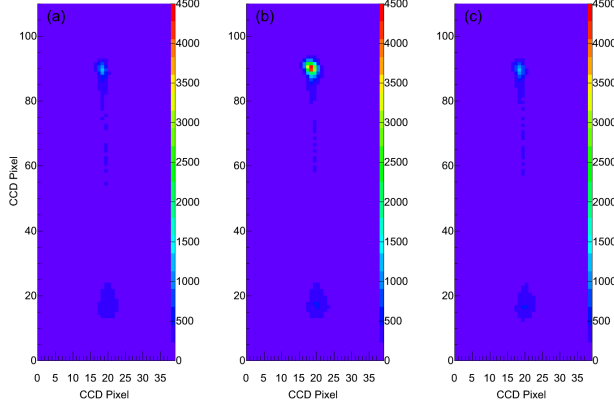
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FIG. 3. Image of a 56-ion deposit with local sXe-only deposits: (a) sXe-only, (b) $\leq$ 56 atoms in sXe, (c) sXe-only. Pixels are 5 μm$\times$5 μm.
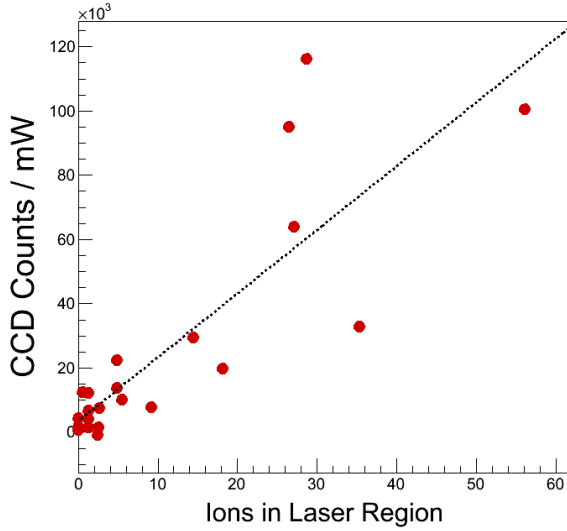

FIG. 4. 619-nm Ba fluorescence vs. number of ions deposited. *Could use better ion beam consistency.*

FIG. 5. Image of a single Ba atom (on average) in a focused w = 2.1 μm laser beam, with 60 s exposure of 0.18 mW laser power.

responding to a single Ba atom is shown in Fig. 5. A solitary peak is observed from the Ba in the laser region.

A Gaussian fit to the images gives a $1/e^2$ radius of about 12 μm, which is much larger than the laser beam radius of w = 2.1 μm. Aberrations and vibrations in the collection optics and imperfections in the surface of the sXe layer could contribute to blurring of the image. Relative motion of the laser and the window could also lead to image broadening and to exposure of more Ba atoms than calculated above. The latter effect has been checked by observ-
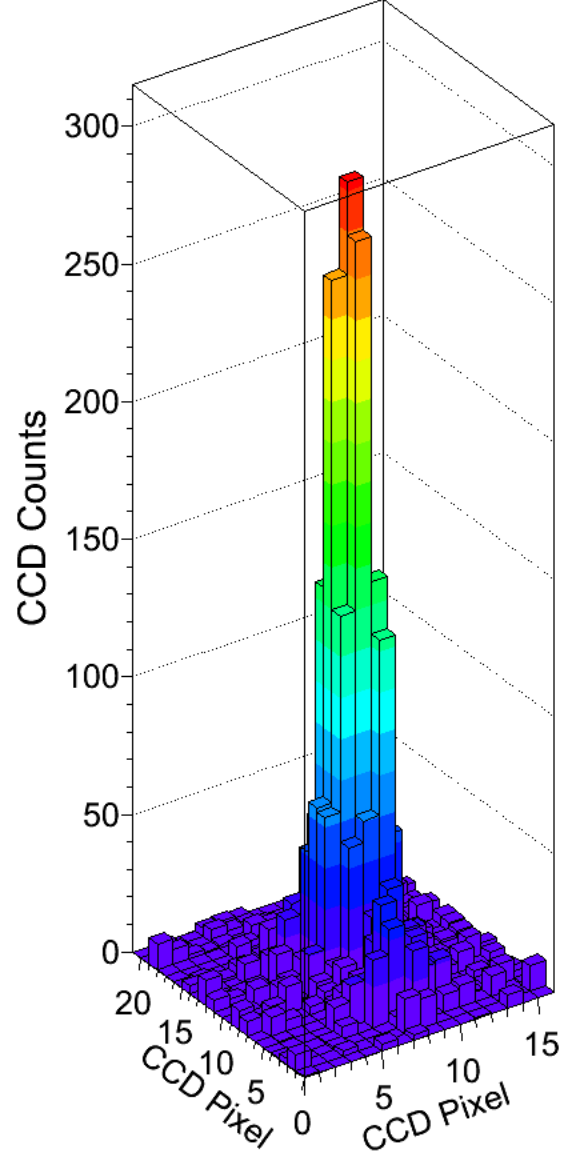
ing images of the focused laser relative to a reference point on the sapphire window on time frames down to 50 ms. Vibrations were observed on the order of 15 μm in x and 4 μm in y, mainly due to cryostat vibrations, which corresponds to a total coverage of about 7 times the laser region. However, the signal at a given time still comes from an area the size of the laser region. Thus, the signals observed correspond to an average number of atoms according to
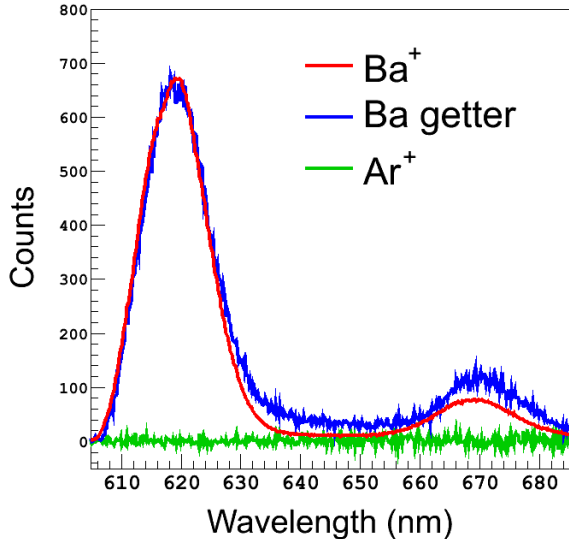
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FIG. 6. Observation of deposits from three different sources in sXe. Curves are scaled due to different deposit sizes.

the laser spot size, though the total number of atoms responsible is about 7 times larger.

A. Scanned Images

Put scanned image of single atoms here.

B. Identification of 619-nm Peak as Ba in sXe

Deposits made with the neutral Ba getter source are compared to a Ba^+ deposit in Fig. 6. Identical spectra are observed using the different sources under similar deposit conditions, confirming the identification of neutral Ba. Another peak at 670 nm which is mentioned in [7] is also attributed to Ba, likely in another Xe matrix site. Observing Ba in the 619-nm matrix site at the low-energy (thermal) deposit from the getter is also good news for the prospect of grabbing Ba/Ba^+ on a probe out of lXe.

An absence of fluorescence is observed from deposits of Ar^+ in sXe (Fig. 6). Ar^+ ions are deposited with the same ion beam, also at 2000 eV, and under the same conditions. This further rules out any matrix-damage-related sources of the 619-nm peak, such as fluorescent color centers.

C. Backgrounds

Very low concentrations of Cr^{3+} in the sapphire bulk (sub-ppb level) produce a broad fluorescence, the tail of which enters the 610-630 nm region passed by the band-pass filter and produces the faintly visible line through the sapphire window discussed above. This fluorescence is identified as Cr^{3+} by its excitation spectrum, identical to that of the well-known lines around 693 nm. *Show figure?* Commercially available c-plane quality sapphire *(mention Meller?)* has sufficiently low concentrations for the 619-nm single atom signal level.

Another background is observed on the surfaces of the window, also caused by laser excitation. This is more of a nuisance, as it not as easily distinguished from the Ba in an image. It also exhibits bleaching, which can cause confusion in subtractions, though its bleaching rate is different from that of the 619-nm Ba emission. *(Mention temp.-dependence?)* To minimize this, the window is exposed to the laser until the background reaches a steady value. The set of deposits used for Fig. 4 was done after about an hour of pre-exposure of the sapphire window to the focused laser.

IV. CONCLUSIONS

The 619-nm emission peak observed in deposits of Ba^+ and Ba in sXe is attributed to neutral Ba *in a stable and relatively abundant matrix site*. Images of 619-nm fluorescence in a focused laser region from Ba atoms are achieved down to an average number of Ba atoms at the single-atom level. Successful detection of Ba atoms in sXe at this level is a significant step toward Ba tagging in nEXO.

ACKNOWLEDGEMENTS

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