

DISSERTATION

IMAGING OF SINGLE BARIUM ATOMS IN SOLID XENON FOR BARIUM TAGGING IN THE
nEXO NEUTRINOLESS DOUBLE BETA DECAY EXPERIMENT

Submitted by

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ABSTRACT

The nEXO experiment is designed to search for zero-neutrino double beta decay of the isotope ^{136}Xe , in order to better understand the nature of neutrinos. Since the daughter of this decay is barium (^{136}Ba), detecting the presence of ^{136}Ba at a decay site (called "barium tagging") provides an additional discriminator to reject backgrounds in the search for this decay. This would involve detecting a single barium ion from within a macroscopic volume of liquid xenon. One proposed barium tagging method is to trap the barium ion in solid xenon (SXe) at the end of a cold probe, and then detect the ion by its fluorescence in the solid xenon. William M. Fairbank Jr.'s group at Colorado State University has been working toward this goal with steady success for some time. In this thesis, I demonstrate successful detection of neutral Ba in SXe down to the single atom level, after deposition from vacuum.

ACKNOWLEDGEMENTS

Bill, Chris for all the help in the lab and building things, Shon and Brian for creating the system and training me (Shon) and for the preliminary results, Adam, Cesar, Kendy.

If you want the Leif thing, uncomment it in csuthesis.cls. It says something like "this dissertation is typeset in ... designed by Leif Anderson.

Jamie and LEW, and other fam.

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CHAPTER 1

INTRODUCTION

Neutrinos have been at the forefront of discovery since their prediction by W. Pauli, who proposed in 1930 the existence of a neutral, unobserved particle to explain the apparent violation of energy conservation in beta decay [1]. He admitted that neutrinos (then deemed “neutrons” – what we now know as neutrons had not been discovered yet either) should be difficult to observe experimentally, but also that it seemed unlikely that they would never have been noticed before. As it turns out, they are much more difficult to observe than he predicted; they will not be noticed without extreme experimental techniques.

A theory formulated in 1933 by E. Fermi for beta decay [2], including the neutrino, would be the beginnings of weak theory, and the development of the very successful Standard Model (SM) of particle physics. However, the SM assumes that neutrinos are massless. The discovery of non-zero neutrino mass via neutrino oscillations in the late 1990s [3] shows that there is more new physics to be discovered – a path to follow in the further unification of physical theory.

Neutrinos are extremely difficult to study, but the rewards are profound. The favored See-saw Mechanism is a theory which predicts the extreme lightness of neutrinos, while also predicting very heavy neutrinos. The existence of heavy neutrinos in the high-energy environment of the early universe, coupled with the possibility of the violation of CP conservation which could slightly favor the production of matter vs. anti-matter in decays of these heavy neutrinos, could explain why the universe exists as we know it. [4]

If this theory is correct, then neutrinos would be described by the Majorana formulation rather than Dirac, and the process of neutrinoless double beta decay ($0\nu\beta\beta$) would be allowed. Observation of $0\nu\beta\beta$ would simultaneously demonstrate that neutrinos are Majorana particles, as well as give a measurement of the absolute mass itself [5]. This chapter outlines the current theory for neutrinos, and then describes the $0\nu\beta\beta$ experiments EXO-200 and nEXO, in order to motivate barium tagging for nEXO.

1.1. NEUTRINOS

Neutrinos are chargeless leptons which only interact via the weak force (and gravity). There are three known “flavors” of neutrinos, each corresponding to one of the three known leptons: ν_e , ν_μ , and ν_τ . These are the eigenstates in the basis of the weak force, so they are the states in which a neutrino will interact via the weak force.

1.1.1. NEUTRINO OSCILLATION AND MASS. The postulate that neutrinos have an energy basis which is different from the flavor basis predicts the phenomenon of oscillation – that the time evolution of an initially pure flavor state (as a neutrino will be produced) will result in a time-dependent probability of measuring the other two flavors as well.

The very small mass of a neutrino (assumed zero in the SM), specifically relative to its momentum, lets one write its Hamiltonian in terms of mass squared differences $\Delta m_{ij}^2 = m_i^2 - m_j^2$, where $i,j = 1,2,3$, referring to what we then call mass states. The mass basis is really the energy basis with the small mass approximation, along with dropping some constant terms in the Hamiltonian (which do not affect time evolution). Writing the time evolution in terms of mass squared differences means that neutrino oscillation experiments can produce measurements of these differences. In fact, the discovery of neutrino oscillation was the first (and only, so far) demonstration that neutrinos have a non-zero mass. Without

neutrino mass (particularly without differences between the masses of the mass states), neutrinos would not oscillate.

Neutrino oscillation experiments also provide measurements on the amount of mixing between the flavor basis and the mass basis. We define the mixing between them by a rotation in terms of three mixing angles, θ_{12} , θ_{23} , and θ_{13} . Transformation between the flavor and mass bases is done with the following unitary matrix, called the Pontecorvo–Maki-Nakagawa-Sakata (PMNS) matrix:

$$(1) \quad U = \begin{pmatrix} 1 & 0 & 0 \\ 0 & c_{23} & s_{23} \\ 0 & -s_{23} & c_{23} \end{pmatrix} \begin{pmatrix} c_{13} & 0 & s_{13}e^{-i\delta} \\ 0 & 1 & 0 \\ -s_{13}e^{i\delta} & 0 & c_{13} \end{pmatrix} \begin{pmatrix} c_{12} & s_{12} & 0 \\ -s_{12} & c_{12} & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & e^{i\alpha_1/2} & 0 \\ 0 & 0 & e^{i\alpha_2/2} \end{pmatrix}$$

$$= \begin{pmatrix} c_{12}c_{13} & s_{12}c_{13} & s_{13}e^{-i\delta} \\ -s_{12}c_{23} - c_{12}s_{23}s_{13}e^{i\delta} & c_{12}c_{23} - s_{12}s_{23}s_{13}e^{i\delta} & s_{23}c_{13} \\ s_{12}s_{23} - c_{12}c_{23}s_{13}e^{i\delta} & -c_{12}s_{23} - s_{12}c_{23}s_{13}e^{i\delta} & c_{23}c_{13} \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & e^{i\alpha_1/2} & 0 \\ 0 & 0 & e^{i\alpha_2/2} \end{pmatrix}$$

where $c_{ij} = \cos \theta_{ij}$ and $s_{ij} = \sin \theta_{ij}$. δ is a phase factor related to lepton CP violation, and α_i are Majorana phases.

Studying oscillations of neutrinos from different kinds of sources, with different energies and path lengths, can isolate sensitivities to the different parameters. For example, the study of solar neutrinos (neutrinos emanating from nuclear fusion reactions in the core of the sun) provides sensitivity to θ_{12} and Δm_{12}^2 . The oscillation parameters so far measured shown in Table 1.1:

TABLE 1.1. Best-fit values for neutrino oscillation parameters, from a global fit to oscillation experiment data. Parameters which depend on the mass hierarchy have values for NH (IH). The atmospheric parameter Δm^2 is defined as $\Delta m^2 = \Delta m^2 = \Delta m_{31}^2 - \Delta m_{21}^2/2 > 0$ ($\Delta m^2 = \Delta m_{32}^2 + \Delta m_{21}^2/2 < 0$). [6]

Parameter	Measurement ($\pm 1\sigma$)
Δm_{12}^2	$7.54^{+0.26}_{-0.22} 10^{-5} \text{ eV}^2$
$ \Delta m^2 $	$2.43 \pm 0.06 (2.38 \pm 0.06) 10^{-3} \text{ eV}^2$
$\sin^2 \theta_{12}$	0.308 ± 0.017
$\sin^2 \theta_{23}$	$4.37^{+0.033}_{-0.023} (4.55^{+0.039}_{-0.031})$
$\sin^2 \theta_{13}$	$0.0234^{+0.0020}_{-0.0019} (0.0240^{+0.0019}_{-0.0022})$
δ/π (2σ range)	$1.39^{+0.38}_{-0.27} (1.31^{+0.29}_{-0.33})$

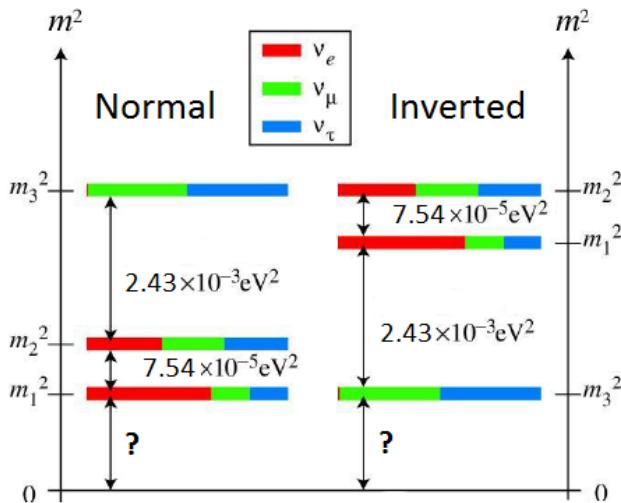


FIGURE 1.1. The two possible hierarchies of neutrino masses. The colors depict the mixing between the mass and flavor bases.

Note that only the absolute value of the atmospheric neutrino oscillation parameter Δm^2 is known. As a consequence, there are two possibilities for the hierarchy of the three neutrino masses. These are called the Normal and Inverted Hierarchies, as shown in Fig. 1.1. The correct mass hierarchy remains unknown, but next-generation neutrino experiments, possibly including nEXO, will be able to discern this.

Neutrino oscillation demonstrates that neutrinos have non-zero mass, and though oscillation experiments measure the mass squared differences, we still do not have a measurement of the absolute masses of the three neutrinos. Cosmology can put limits on the sum of the



FIGURE 1.2. Two-neutrino (left) and neutrinoless (right) double beta decay.

three neutrino masses. The Planck collaboration reports an upper bound on this sum at $\sum_i m_i < 0.23$ eV [7]. The KATRIN experiment will search for absolute neutrino mass with limiting capability of $m_{\bar{\nu}_e} < 0.2$ eV (90% CL) by measuring the spectrum of tritium beta decay near the Q-value, searching for deviation from the spectrum with zero neutrino mass [8].

1.2. DOUBLE BETA DECAY

Double beta decay is the simultaneous decay of two neutrons in a nucleus into two protons and two electrons. Two-neutrino double beta decay ($2\nu\beta\beta$), shown in Fig. 1.2(left), is allowed by the Standard Model and has been observed in eleven isotopes with half-lives between 10^{19} and 10^{21} years. Similar to beta decay, a neutrino accompanies each electron in this decay, broadening the spectrum of the summed electron energy. This is a second-order process, making it a rare decay, and requiring low backgrounds to measure.

$0\nu\beta\beta$, shown in Fig. 1.2(right), is a postulated mode of double beta decay. In this case, the neutrino is exchanged as a virtual particle (which would require that it is a Majorana

particle), and there are no neutrinos in the final products. If discovered, not only would neutrinos be determined Majorana particles, but their absolute mass could also be measured as it relates to the $0\nu\beta\beta$ half-life according to Eqn. 2:

$$(2) \quad T_{1/2}^{0\nu} = (G^{0\nu}(Q, Z)|M^{0\nu}|^2 \langle m_\nu \rangle^2)^{-1}$$

where $T_{1/2}^{0\nu}$ is the $0\nu\beta\beta$ half-life, $G^{0\nu}$ is a known phase space factor, and $M^{0\nu}$ is a model-dependent nuclear matrix element. The effective electron neutrino mass $\langle m_\nu \rangle$ is the expectation value of the mass for a pure electron neutrino:

$$(3) \quad \langle m_\nu \rangle = \sum_i U_{ei}^2 m_i.$$

The sum of the energies of the emitted electrons in double beta decay will serve as the distinction between the two-neutrino and zero-neutrino modes, shown in Fig. 1.3. In the two-neutrino mode, the total decay energy is shared probabilistically between the electrons and the neutrinos (the nucleus recoil energy is negligible), resulting in a broad distribution in the summed electron energy. (Recall the similarly broad electron energy in single beta decay, which ultimately led to discovery of the neutrino involved.) But in the zero-neutrino mode, all of the decay energy is carried away by the two electrons, resulting in only a single allowed value for the summed electron energy – a peak in the summed electron energy spectrum at the Q-value.



FIGURE 1.3. Conceptual two-neutrino (blue) and zero-neutrino (red) double beta decay spectra.

The rarity of double beta decay requires very low backgrounds, especially around the Q-value for the $0\nu\beta\beta$ search. The next sections describe the experiments EXO-200 and its next-generation successor, nEXO.

1.3. ENRICHED XENON OBSERVATORY

EXO-200 and nEXO (EXO standing for Enriched Xenon Observatory) are a progression of two experiments, each a LXe time projection chamber (TPC) designed to study the double beta decay of the isotope ^{136}Xe , and ultimately to search for the zero-neutrino mode. Xe is fairly unique among the double beta decay isotopes in that it can be studied in a gas or liquid TPC instead of solid crystals or foils. The 3D event position reconstruction abilities of a TPC have advantages in background reduction, described in section 1.3.1. Purification of Xe is straightforward and can be done continuously in a detector. LXe is transparent, and produces substantial ionization and scintillation at 178 nm when energy is deposited in the LXe [9]. A liquid TPC approach also offers the opportunity to identify, or “tag”, the

daughter Ba⁺⁺ at the site of the double beta decay event, which would provide a background-free identification of $0\nu\beta\beta$ [10]. Barium tagging is the focus of our group at CSU and is the subject of this thesis. The following sections describe the EXO-200 experiment, as well as nEXO, the next-generation tonne-scale LXe TPC which is now in the design stages. EXO-200 does not have barium tagging implemented, but it is hoped that nEXO will.

1.3.1. EXO-200. EXO-200 has been operational since 2011. It is a TPC using 110 kg of active LXe enriched to $80.672\% \pm 0.14\%$ ^{136}Xe [9], designed to probe Majorana neutrino masses down to around 109-135 meV (the range covers different matrix element calculations) [11], and is located about half a mile underground in the Waste Isolation Pilot Plant (WIPP) near Carlsbad, NM. This mine is in a salt basin, which contains lower levels of Uranium and Thorium than a more typical mine in rock.

A schematic of the TPC in the class 100 cleanroom is shown in Fig. 1.4. Several layers of lead wall surround the copper cryostat, which is filled with HFE-7000, a cryogenic fluid which keeps the TPC cooled to LXe temperatures, as well as aids in shielding. The TPC vessel is made of low-radioactivity copper, and is kept as thin as possible to minimize backgrounds. Scintillating panels on the outside of the cleanroom provide muon veto.

A cut-away view of the EXO-200 detector is shown in Fig. 1.5. It is two mirrored TPCs which share a cathode. The detection planes are a combination of ionized charge induction/collection wires and large area avalanche photodiodes (LAAPDs) which detect scintillation light [12].



FIGURE 1.4. Drawing of cleanroom laboratory in the WIPP drift.

A photograph of the detection plane is shown in Fig. 1.6, and a schematic of event detection is shown in Fig. 1.7. When a double beta decay event occurs in the LXe, the energetic electrons ionize many surrounding Xe atoms, as well as produce a scintillation signal. Scintillation light provides a time stamp for the event as well as an energy measurement.

The cathode is set to -8 kV, providing an electric field of 374 V/cm across the 20 cm drift length of each TPC. Ionized electrons drift from the decay site, first passing the v-wires, which receive an induction signal, and are then collected by the u-wires, which are set at a 60° angle from the v-wires. An electric field of 778 V/cm between the u- and v-wires ensures 100% v-wire transparency. The charge collection provides an energy measurement complimentary to that of the scintillation. Together, the u- and v-wires give an x/y position measurement for the event. The time between the initial scintillation detection and the charge collection give a



FIGURE 1.5. EXO-200 TPC cutaway [9].

z position, and a 3D position can be reconstructed for the event. [9] Additional scintillation resulting from recombination of ionized electrons leads to a well-known microscopic anti-correlation between ionization and scintillation signals [13]. Applying this to the combination of those signals improves the energy resolution. This correction defines a combined energy axis, called the rotated energy. Energy resolution is important in a $0\nu\beta\beta$ search, as it distinguishes those events from $2\nu\beta\beta$ events in the tail of their spectrum.



FIGURE 1.6. View of the detection plane in one of the two EXO-200 TPCs.



FIGURE 1.7. EXO-200 event detection.

Having a reconstructed 3D event position is important in several ways. Firstly, position-based corrections on scintillation and charge collection can be applied. For charge, electronegative impurities in the LXe will absorb the drifting charge, requiring a drift-length (z-position) correction. High purity levels, measured in terms of electron lifetime, of $2\text{--}3 \times 10^3 \mu\text{s}$ are maintained in EXO-200, resulting in a small correction of few % for maximal drift lengths. For scintillation, a 3D correction is applied, as some regions have more efficient light collection by the LAAPDs. A 3D position also allows a fiducial volume to be defined. The effect of radio-impurities on detector surfaces, e.g. radon daughters collecting on the cathode, is mitigated by defining a stand-off distance required for events used in analysis. Finally, 3D reconstruction allows the distinction between single-site (SS) and multi-site (MS) events. A MS event is one where two spatially separated events occur in the same $2048\text{-}\mu\text{s}$ time window. These are mostly caused by gamma rays interacting in the LXe, which can Compton-scatter several times. Rejecting MS events strongly separates gamma events from double beta decay events. [9] Of course, barium tagging will also require a 3D reconstructed position in nEXO.

The final data set is fit using a combination of probability distribution functions (PDFs) for $0\nu\beta\beta$, $2\nu\beta\beta$, and all possible backgrounds. Fits to the final energy spectrum data for (a) SS events, and (b) MS events are shown in Fig. 1.8 for the most recent $0\nu\beta\beta$ search analysis. The green bands beneath each plot show the residuals vs. energy. The $2\nu\beta\beta$ spectrum, in gray, dominates the backgrounds in the SS spectrum. The vertical red lines in the SS spectrum outline the $\pm 2\sigma$ region of interest around the Q-value, where the $0\nu\beta\beta$ peak will lie. The insets are a zoom into this region. The fit value for $0\nu\beta\beta$ in this dataset is non-zero, but it is consistent with the null hypothesis at 1.2σ . This sets an upper limit on the



FIGURE 1.8. EXO-200 energy spectrum from $0\nu\beta\beta$ search analysis [14].

$0\nu\beta\beta$ half-life at $T_{1/2}^{0\nu\beta\beta} < 1.1 \times 10^{25}$ yr (90% CL), which corresponds to $\langle m_{\nu_e} \rangle < 190\text{-}450$ meV depending on nuclear matrix element calculations [14]. EXO-200 reports the $2\nu\beta\beta$ half-life measurement in [9] of $T_{1/2}^{2\nu\beta\beta} = 2.165 \pm 0.016(\text{stat}) \pm 0.059(\text{sys}) \times 10^{21}$ yr. This is consistent with previous measurements by EXO-200 in 2011 [15] and KamLAND-ZEN in 2012 [16].

1.3.2. nEXO. The next-generation successor to EXO-200 is nEXO, a tonne-scale LXe TPC which will probe Majorana neutrino masses down to the 10 meV scale. The sensitivity projections for nEXO are shown in Fig. 1.9, along with those of EXO-200. nEXO will reach phase space where the two possible mass hierarchies begin to split; if nEXO successfully



FIGURE 1.9. nEXO projected sensitivity to the Majorana neutrino mass vs. the minimum of the three masses m_{min} . [check for updated version](#)

observes $0\nu\beta\beta$ in these regions, it may be able to also determine the mass hierarchy. Barium tagging will push the sensitivity further into the region allowed only by the normal hierarchy [ref for this?].

A schematic of the experimental setup is shown in Fig. 1.10 in one of the possible locations for nEXO, the SNOLAB cryopit. Similar to EXO-200, the copper-housed TPC will be submerged in HFE fluid, inside a copper cryostat. The cryostat will be insulated and submerged in a large volume of water shielding, in which photo-multiplier tubes could

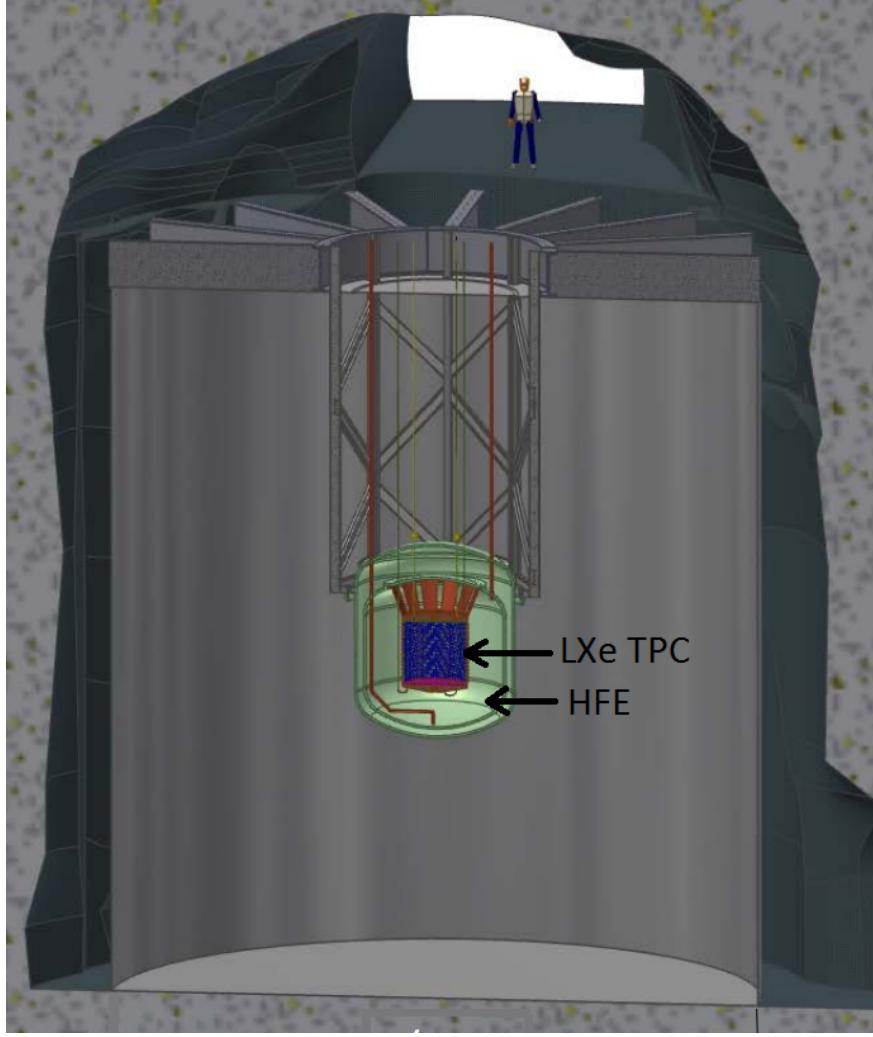


FIGURE 1.10. nEXO TPC in the SNOLAB cryopit.

provide muon veto by observing Cherenkov radiation. nEXO will be a single TPC with charge and light readouts at opposing ends of the TPC. Rather than wires, nEXO will use tile electrodes for charge readout, shown in Fig. 1.11.

1.3.3. BARIUM TAGGING. The backgrounds observed around the Q-value in EXO-200 can be expected to scale up with the size of nEXO. But with the veto power of barium tagging, nEXO's sensitivity to $0\nu\beta\beta$ scales with the total ^{136}Xe mass in the detector, vs. the square root of that mass without barium tagging.



FIGURE 1.11. nEXO readout.

Several possible barium tagging techniques have been proposed. Perhaps the most natural concept is to direct one or more lasers at the decay site to induce fluorescence of the barium daughter. This technique was explored thoroughly by Fairbank's group, and was abandoned when reliable fluorescence of Ba^+ in LXe could not be observed [ref Kendy's thesis, after kamland in list].

Three barium tagging techniques continue to be explored. One of these is to grab the daughter on a surface, brought to the decay site by a probe, and then move it to a location where it can be desorbed from that surface by an infrared laser, and subsequently resonantly ionized by two lasers in order to detect it by time-of-flight spectroscopy. The apparatus for the study of this method is described in [17], along with some initial results. Another concept is to also grab the daughter on the surface of a probe, but then to thermally desorb it for subsequent trapping and detection [ref upcoming Liang paper, after Twelker].

The other remaining technique for barium tagging, now the concentration of Fairbank's group and the subject of this thesis, is called tagging in SXe. Here, a cold probe would go to the site of the candidate $0\nu\beta\beta$ event in order to trap the Ba/Ba^+ in a small amount of SXe,

and then observe it by its laser-induced fluorescence in the SXe, a technique called matrix isolation spectroscopy.

A concept for a probe is shown in Fig. [ref fig cryoprobe]. The SXe forms on a sapphire window or plug. Sapphire is a good candidate for a substrate; it has good thermal conductivity at low temperature, and is extremely transparent. An excitation laser would be brought into the probe through a fiber, and aimed through the sapphire to excite the Ba/Ba⁺ in the SXe. A return fiber could collect the laser reflections to measure the SXe thickness via interference fringes. The Ba/Ba⁺ fluorescence would then be collected by a lens/filter system and imaged onto a CCD. Additional components, not shown, would be required for cooling the sapphire, either by liquid He or by a Joule-Thompson nozzle, as well as a thermometer.

Whether the daughter Ba will neutralize or remain ionized is not yet known. It is however expected that a Ba⁺⁺ ion will neutralize once to Ba⁺ in lXe, since the lXe conduction band gap is slightly less than the ionization potential for Ba⁺ [10]. Further neutralization could also occur. A study of neutralization of alpha decay daughters in EXO-200 has observed a fairly equal balance of neutralized daughters vs. ionized daughters [ref alphaion]. The feasibility of detecting single Ba ions as well as atoms is still of interest.



FIGURE 1.12. Concept for a cryoprobe containing excitation/collection optics.

CHAPTER 2

THEORY

2.1. BA/BA⁺ SPECTROSCOPY IN VACUUM

The lowest-lying energy levels in vacuum for Ba and Ba⁺ are shown in Fig. 2.1. For Ba, the main transition is between the ground $6s^2 \ ^1S_0$ to the excited $6s6p \ ^1P_1$ state. Spin(?)-suppressed transitions between the P state and three metastable D states results in a decay in to a D state after about 350 excitations. For Ba⁺, two strong transitions exist between the ground $6s \ ^2S_{1/2}$ and the $6p \ ^2P_{1/2}$ and $6p \ ^2P_{3/2}$ excited states. Transitions to the two metastable D states are higher than for the atom, resulting in a decay into a D state after about 4 excitations.

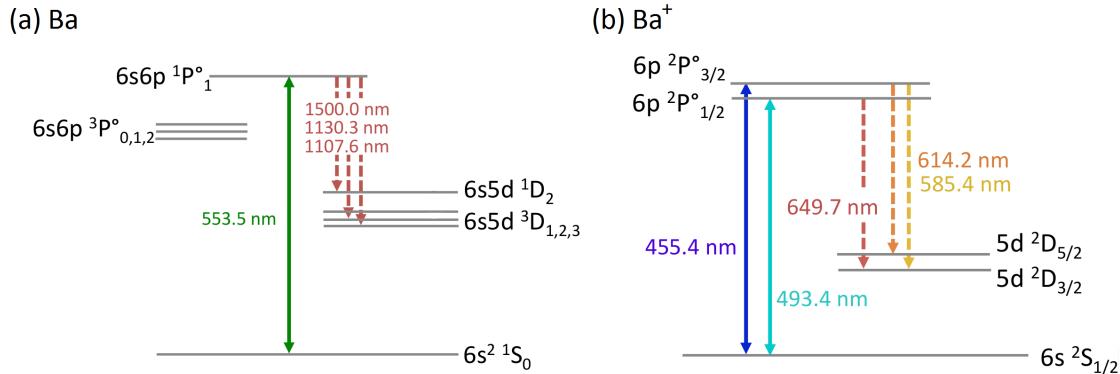


FIGURE 2.1. Energy level diagrams for (a) Ba and (b) Ba⁺ ... *bah, you should have the blue AND the red Ba lines in there for reference from re-pumping section.*

These energy levels and their transition rates are well known, and are documented in the NIST Atomic Spectra Database. Single atom/ion detection by spectroscopy generally requires, in addition to the main excitation laser, lasers to provide transitions out of the metastable D states once the atom/ion decays into one of them. For the atom, this requires three additional infrared lasers. Single atom trapping/detection [do they actually have

single-atom in that paper? ... also they did it with a red laser too] in a magneto-optical trap (MOT) is achieved in [20]. Single Ba⁺ observation requires only two lasers if the ²P_{1/2} excited state is used. This is demonstrated in [ref something that does this ... is there something? How about the Carleton group? I think MOE has a reference to it maybe – yeah, its [19]].

2.2. MATRIX ISOLATION SPECTROSCOPY

In the solid matrix, the Ba/Ba⁺ spectroscopy is affected by its interaction with neighboring Xe atoms.

Kind of a main idea is the very different interaction between M and G for the ground vs. excited state. This leads to Franck-Condon rule and to Jahn-Teller. Shon doesn't really say that, I don't think. May just say that it's a van der waal force, w/o writing the force terms, though Crepin doesn't even seem to mention that as important.

May be good matrix isolation theory references in Ba Spec... yeah, 1 and 2 i think ... shon's 55, 57, 58 are probably good too and maybe overlapping

Does the lead into matrix isolation in Crepin start with just the interaction w/ noble gas? You could have a section for that like Shon, or just have it within this section.

The spectroscopy of a species trapped in an inert solid matrix is called matrix isolation spectroscopy, the concept of which was pioneered around 19?? by [that one guy] [ref ... maybe [1] of ba spec]. Though absorption and emission are significantly broadened and shifted, a species can retain properties of its vacuum counterpart, such as quantum numbers.

Mention optimal guest:host ratio? This number will be referred again to in Results: search Results for keyword barfbreath

Studies of some matrix isolation systems have been made. The most thoroughly studied has been Na(?) in solid matrices of (Ar, Kr, Xe ?)... (Na? Mg?)...[refs]

We are of course interested in the spectroscopy of Ba and/or Ba⁺ in SXe matrices, and this particular system has not been studied until recently. The first report of the spectroscopy of neutral Ba in SXe, along with candidate fluorescence peaks for Ba⁺ in SXe, was published by our group in [ref ba spec], and conversations following this publication with ???'s group in ??? have confirmed our basic observations of the absorption spectrum of Ba in SXe.

From here forward I will refer to the host species as Xe and the guest as Ba/Ba⁺, specific to our system. The leading interaction between [put this here if the van der waals is only necessarily the force for Ba in particular] the Ba/Ba⁺ and a neighboring Xe atom is a (the) Van Der Waals (sp?) force, an induced dipole-dipole interaction. Xe is the most polarizable (sp?) of the noble gases.

The forms for this force(?) is shown in Eqn. [ref van der ba] for Ba and Eqn. [ref van der ba+] for Ba⁺. Talk about it.

This binding between the Ba/Ba⁺ and its surrounding Xe atoms results in vibrational modes, and the Franck Condon (sp?) principle applies. Fig. [ref fig franckcondon] helps illustrate this effect. In a cold matrix, the system will be in the ground vibrational state before excitation. The distribution of the wavefunction, even for the ground state alone, overlaps in space in general with more than one of the excited state vibrational modes, resulting in a broadening in energy of the absorption.

Rapid decay occurs to the lowest vibrational mode in the excited state before electronic decay can occur [ref] **[is this just for Ba?]**. Then a similar broadening in emission energy occurs as several overlapping vibrational mode wavefunctions exist in the ground electronic

state, and a redshift is also observed as some energy was dissipated into phonons in the crystal in vibrational mode decays. ([How can you get a blueshift?](#))

Shifts and broadening in absorption and emission depend in general on the distribution of Xe atoms around the Ba/Ba⁺. The fcc crystal structure of the Xe restricts these environments to discrete number of so-called matrix sites, defined by (a) whether the Ba/Ba⁺ is [intersituational or whatever – is that still a thing?], and (b) the number of vacancies in the Xe matrix surrounding the Ba/Ba⁺. Experimental observation of emission peaks from different matrix sites of [Na] in solid noble gases is reported in [ref(s)], with theoretical calculations in [ref(s)] attributing observed peaks to specific vacancy distributions defining the matrix sites (maybe this has been done?).

Energy level transition probabilities can also be affected in a matrix. Distortions in electron wavefunction shapes by asymmetric matrix sites can affect radiative transitions by altering parity [ref]. If electronic potential energy curves cross each other, nonradiative transitions can also become allowed for otherwise forbidden transitions [ref]. ([Is a phonon emission called a nonradiative transition / does this actually happen?](#))

Jahn-Teller?

The detectability of a single atom/ion can depend greatly on altered transition rates. For example, in the Ba atom, matrix-allowed decay of the ¹P₁ to the ³P states could be much stronger than the main transition back to ground, which would suppress fluorescence and make single-atom detection impossible. But the matrix could also help by strengthening decays from the D states back to ground, eliminating any need for re-pump lasers to keep the transition cycle going.

2.2.1. 6-LEVEL SYSTEM. Neutral Ba in vacuum is only a 5-level system. However, solving a 6-level system is helpful in modeling matrix-isolated Ba where interactions could allow a transition into a 3P state.

2.3. SHON HAD A FLU EFF SECTION

CHAPTER 3

EXPERIMENTAL

Probably need to mention CCD digitaztion pedestal and noise, and dark counts which are zero when cold – did you do that?.

This chapter describes the apparatus at Colorado State University which has been used for all described studies of Ba/Ba⁺ fluorescence in SXe after deposition in vacuum. Our main barium source, a Ba⁺ ion beam, is first described, as well as a purely Ba neutral source. The co-deposit of Ba/Ba⁺ with Xe gas onto a cold sapphire window, subsequent laser excitation, and finally the collection optics for the fluorescence are described.

3.1. ION BEAM

The full ion beam is shown in Fig. 3.1. This is a clean source of Ba⁺ which can do a very wide range of deposit sizes, from billions of ions in a focused laser region all the way down to the single-ion level, and even deposit sparsely enough to scan a focused laser over spatially separated ions.

3.1.1. ION SOURCE. Ba⁺ ions are produced in a Colutron ion gun system [21]. The source is shown in Fig. 3.2. A solid barium charge is placed into the hollowed end of a stainless steel rod, which is then inserted into the discharge chamber, near the hot filament. The barium vaporizes, and escapes the hollowed rod around a loosely threaded set screw. The source is designed to produce a discharge between the anode plate and the filament cathode, through an argon buffer gas leaked into the source chamber. This controlled discharge would then also ionize atoms from the solid charge to produce the desired ion beam, and Ar ions would be filtered out. However, to avoid contamination of the SXe matrix with residual

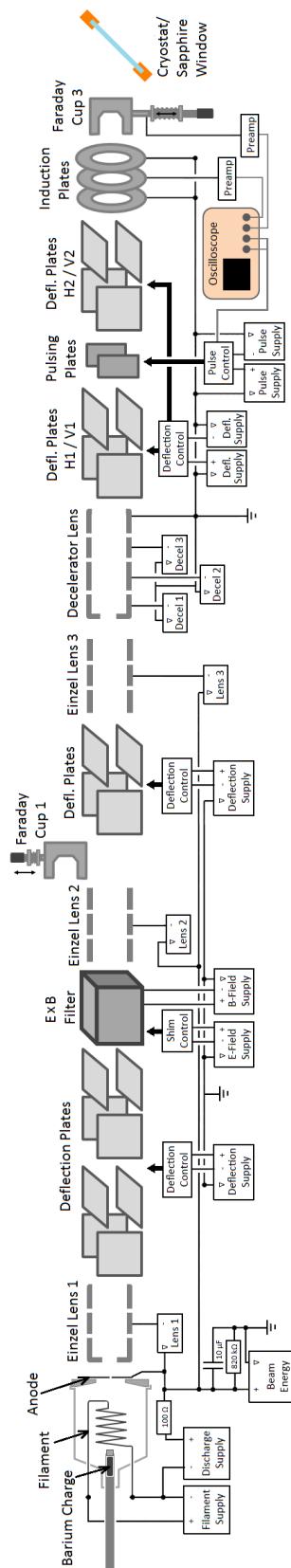


FIGURE 3.1. Ba^+ ion beam.



FIGURE 3.2. Ba^+/Ar^+ ion source.

Ar gas, the buffer gas is not used in this work. We are still able to maintain a discharge between the filament and anode circuits. The longevity of ion current from a single charge (at least several 10s of hours) suggests that Ba is coating the inner walls of the chamber and is depleted slowly. This is supported by the observation of white oxidation of the inner source parts after a few minutes of exposure to air when opening the system. The discharge produces a plasma, containing barium ions, which escapes the chamber through a small hole in the anode, where it enters the acceleration region. The acceleration potential is 2 kV, between the ion source anode and an aperture, which is the first element of Einzel lens 1. This lens approximately collimates the ion beam for passage through the $E \times B$ velocity filter.

3.1.2. $E \times B$ VELOCITY FILTER. The $E \times B$ velocity filter selects Ba^+ by creating perpendicular electric and magnetic fields, which produce opposing forces on charged particles moving straight through the filter. The opposing forces will be equal for ions with velocity

$v = \frac{E}{B}$. Since ion velocity is determined by mass (m), charge (q) and beam potential (V), the filter selects ions satisfying Eq. 4:

$$(4) \quad \frac{m}{q} = \frac{2VB^2}{E^2}$$

where B and E are the magnetic and electric fields, respectively. Those fields are chosen such that the forces are equal for Ba^+ . Other ions will be deflected.

The $E \times B$ filter is shown in Fig. 3.3. Electromagnets provide the vertical magnetic field. Electrode plates and field-shaping guard rings provide the horizontal electric field. The guard rings prevent a lensing and astigmatism effect from fringe fields of the plates [21].

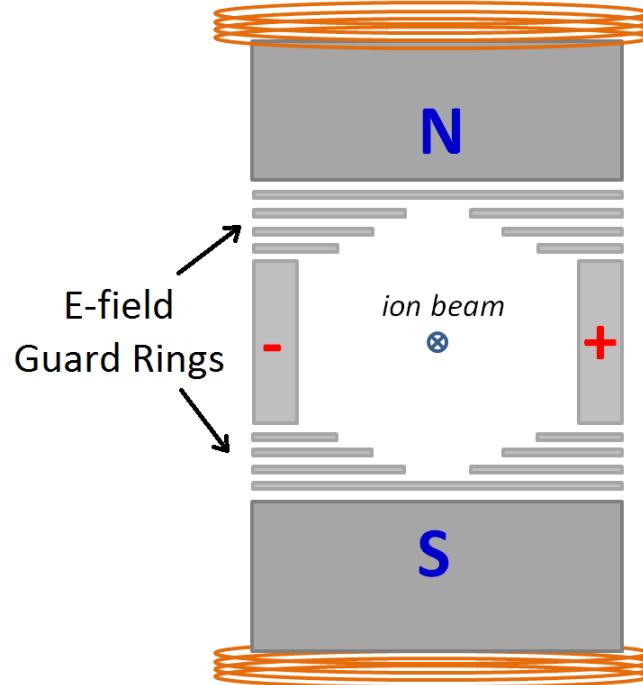


FIGURE 3.3. Colutron $E \times B$ ion velocity filter.

3.1.3. OTHER BEAM COMPONENTS. The first three sets of deflection plates can be used for beam diagnostics, and are set to 0 V during normal operation. The deflection plates just

before the pulsing plates, H1 and V1, are set to constant values of 50 V and 0 V, respectively, which have been selected such that the beam, in both pulsing and continuous modes, can be deposited at the sapphire window for reasonable settings on the final deflection plates, H2 and V2. As described in 3.1.5, different settings in H2/V2 are required for peak ion current in Faraday cup 3 vs. peak deposit at the window.

Einzel lens 2 focuses the beam to pass through the aperture in the first element of the decelerator lens. Einzel lens 3 is not used in this setup. The decelerator lens can be used to vary Ba^+ deposit energy, but in this work it acts as an Einzel lens with only the second element at voltage, and it focuses the beam at Faraday cup 3 (there is no Faraday cup 2 in this setup). Faraday cup 3 measures the ion current during experiments, and is retracted when deposits are being made. Its use in deposit calibration is described in 3.1.5. Faraday cup 1 is used for beam diagnostics, and is usually retracted on its bellows.

3.1.4. ION BEAM PULSING. When running in pulsing mode, the pulsing plates are first set to 200 V and -200 V to deflect the beam, and are pulsed to 0 V for 1 μs for each pulse. The pulsing circuit is shown in Fig. 3.4. Square waves, triggered by LabVIEW at 500 Hz, enter the circuit at (a). The transformed pulse triggers the MOSFET switch, which closes the circuit for the period of the pulse.

The induction plates observe pulses during a deposit. Pulses just prior to a deposit can be observed by cup 3 (as well as the induction plates) for a local measurement of ion current in the pulses. eV Products pre-amplifiers convert the ion current to voltage signals which are read out with a digital oscilloscope. An example of an oscilloscope readout of 16 averaged pulses is shown in Fig. 3.5(left). The pre-amp circuit is solved to retrieve the original current, given by Eqn. 5:



FIGURE 3.4. Pulsing circuit. $R_1 = R_2 = 470 \Omega$, $R_3 = R_4 = 20 \text{ k}\Omega$, $C_1 = C_2 = 680 \text{ nF}$. [22]

$$(5) \quad I = \frac{-(V_{out} + R_1 C \frac{dV_{out}}{dt})}{R_1 M}.$$

$R_1 C$ and $R_1 M$ are determined for each pre-amp. The time constant $R_1 C$ is determined by fitting an exponential to the decay of a signal, and $R_1 M$ is a constant which is then determined by fitting this shaped signal to a known input square pulse. Final shaped currents for signals from the induction plates and cup 3 are shown in Fig. 3.5(right).

Pulsing data also provides confirmation that the beam is composed of Ba^+ . The time between the center of the pulsing plate voltage overlap and the center of the pulse measured by the Faraday cup, along with a measurement of the distance traveled, provides a velocity measurement of the ions. This distance was measured to be $31.5 \pm 0.5 \text{ cm}$, and time-of-flight



FIGURE 3.5. Raw (left) and shaped (right) pulse signals from induction plates and cup 3. The raw induction signal appears small because it is a less sensitive pre-amp (accounted for in shaping).

data, e.g. Fig. 3.6, give 39.8 ± 3.4 amu for Ar^+ and 136.8 ± 6.3 amu for Ba^+ , including an uncertainty on the time of flight of $\pm 0.1 \mu\text{s}$. This rules out any BaO^+ contribution to the Ba^+ ion beam.

3.1.5. CALIBRATION OF ION DEPOSIT. To calibrate the signal at cup 3 to an ion density at the sapphire window, another Faraday cup (cup w) is attached to the cold finger in place of the sapphire window. Firstly, the ratio in $\frac{fC}{pulse}$ between cup 3 and cup w is measured ($\equiv f$). Then, knowing the radius of cup w lets one determine the ion density per pulse at the sapphire window:

$$(6) \quad \frac{ions}{pulse \times m^2} = \frac{Cf}{Ae}$$

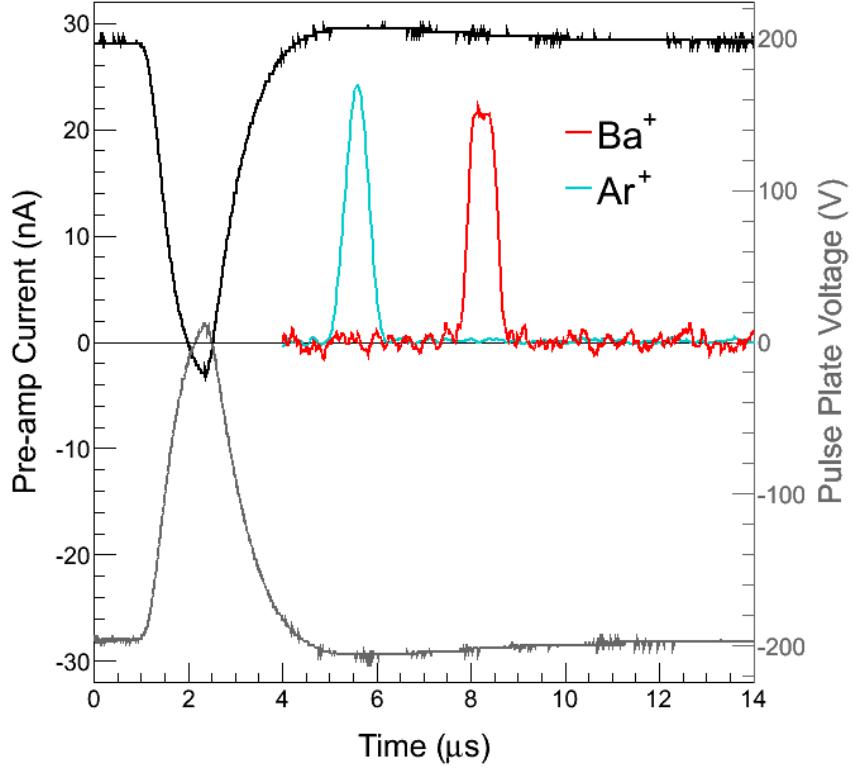


FIGURE 3.6. Arrival time of pulses at cup 3 vs. time of pulsing plate signal (black (+) and gray (-)) for Ar^+ and Ba^+ .

where C is $\frac{fC}{pulse}$ at cup 3, A is the area of cup w, and e is the elementary charge. The required settings of the final deflection plates H2 and V2 are also determined by cup w. These typically differ from the peak values for cup 3 by about 70 V in H2 and 60 V in V2, corresponding to about 4 mm in x and y position at cup w.

3.2. BA GETTER SOURCE

A BaAl_4 getter is employed as a purely neutral Ba source which can be inserted on a bellows to emit toward the sapphire window, shown in Fig. 3.7. Getters were used extensively in previous work [23] for measuring the absorption of Ba in SXe with large Ba deposits, and in identifying emission peaks. A getter is used briefly in this work in identifying the 619-nm fluorescence peak, as described in 4.3.4. Getters used in [23] were exothermic $\text{BaAl}_4\text{-Ni}$



FIGURE 3.7. Apparatus around sapphire window, including Ba getter, and optics for excitation and collection.

flash getters. The getter used here is an endothermic type, designed for more controlled Ba emission.

3.3. SAMPLE DEPOSITION

The Ba^+/Ba is co-deposited with ultra-pure Xe gas onto a cold sapphire window. Sapphire has good thermal conductivity at low temperature and good optical transparency in the visible. The window is held to a coldfinger and is tilted at 45° to allow access of the ion beam as well as the excitation laser and collection optics. To begin a deposit, Xe gas



FIGURE 3.8. Setup for measuring SXe growth rate by interference fringes.

is flowed toward the window via a leak valve. Cup 3 is then retracted to clear the path for Ba^+ ions, which land in the SXe matrix as it grows. Cup 3 is then replaced, and the Xe leak stopped.

SXE matrix growth rate can be measured by interference fringes in a laser reflected against the front surface of the sapphire window, as shown in Fig. 3.8. Fringes for SXe deposition at 52 K and 11 K are shown in Fig. 3.9 for the leak rate used in this work. The refractive index of SXe has a negligible dependence on temperature between 50 and 30 K [24], so these can be compared directly. A somewhat lower rate is observed at 52 K, at 31 nm/s, vs. 37 nm/s at 11 K. To evaporate a sample, the window is heated to 100 K. Fringes appear during this process as well. The full set of fringes for a deposit at 52 K and its evaporation when heated is shown in Fig. 3.10, along with the window temperature. This shows that the SXe evaporates between 73 K and 78 K. Notice that the same number of fringes appear in the deposit and the evaporation.

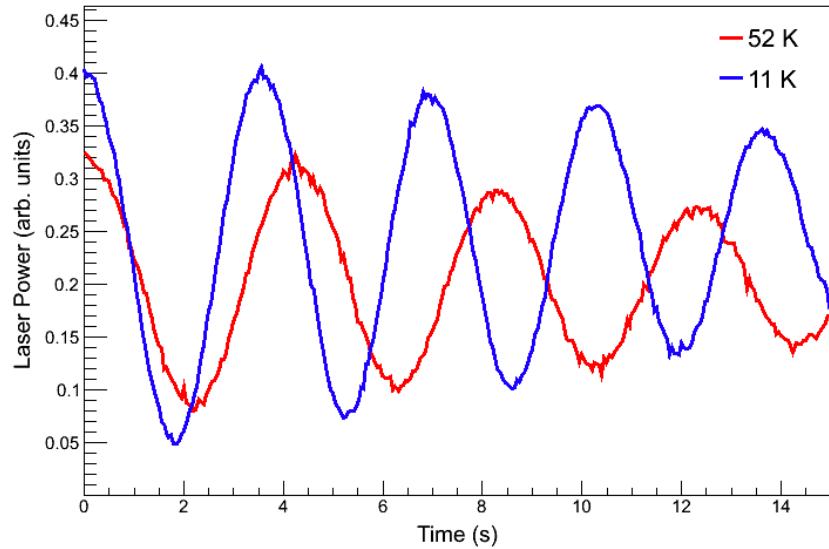


FIGURE 3.9. Interference fringes for the same Xe gas leak rate deposited on the sapphire window at 11 K and at 52 K.

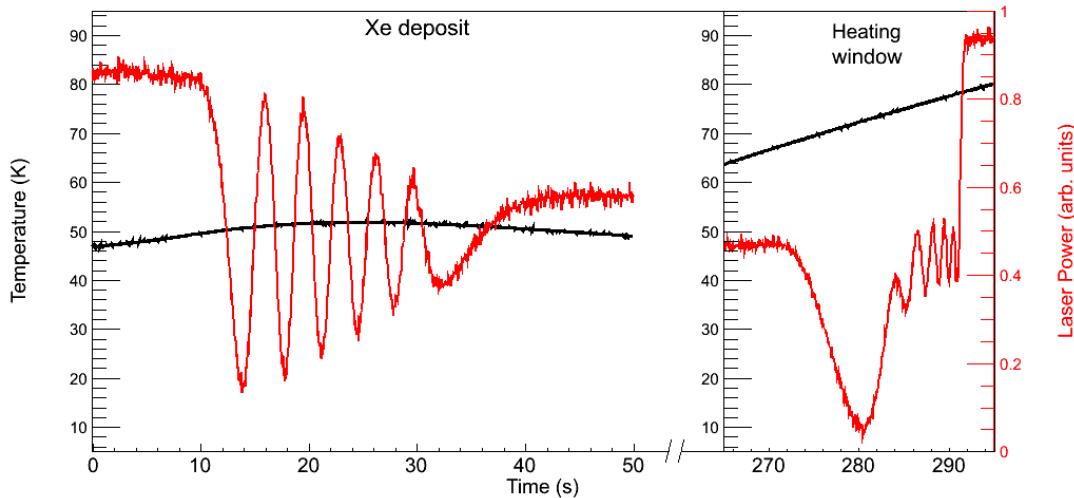


FIGURE 3.10. Interference fringes of a deposit at 52 K and of its subsequent evaporation when heating the sapphire window.

3.4. LASER EXCITATION

Green/yellow laser excitation is done with a Coherent 599 dye laser, pumped by the 514-nm line of a Lexel 3500 Ar ion laser. Rhodamine 110 (R110) dye tunes over 542 - 566 nm, and Rhodamine 6G (R6G) tunes over 567 - 590 nm. Another Coherent 599 contains Coumarin 480 (C480) dye for blue excitation, which is pumped by a Kr ion laser.

A 7.9-cm focal length aspherical lens focuses the laser into the cryostat from the side (Fig. 3.7). This lens reduces spherical aberration in the laser focus so that it can reach the diffraction limit. However, to reach the diffraction limit in the SXe, astigmatism introduced by the tilted sapphire window must be corrected. To do this, compensating astigmatism is introduced by a fused silica optical flat of 1 cm thickness, placed after the lens, tilted in the y-plane, whereas the sapphire window is tilted in the x-plane. The proper angle for the compensator is determined to be about 10° from normal by a ray matrix equation for a sapphire window of thickness 0.5 mm [25]. The effect of the SXe layer is negligible since its thickness is less than a wavelength. With the compensator, overlapping minimum spot sizes of $2.06 \mu\text{m}$ and $2.66 \mu\text{m}$ are calculated for x and y, respectively, corresponding to an effective minimum spot size (w_0) of $2.36 \mu\text{m}$ in the SXe. *Show astig. corr. data w/ 2D gaus fits.*

3.5. COLLECTION OPTICS

Fluorescence is collected above the cryostat (Fig. 3.7). A 50 mm Nikon camera lens collimates the light, and a fluorescence filter sits on top of it. A band-pass filter is used for imaging, and a Raman filter is used for spectroscopy. The fluorescence then reflects off two steering mirrors, and is imaged by a 200 mm Nikon camera lens onto a Roper Scientific liquid-nitrogen-cooled CCD, with a magnification of 4.

The CCD has a removable Princeton Instruments imaging spectrometer. With this attached, the 200 mm camera lens focuses the light onto an inlet slit, which is then imaged by the spectrometer onto the CCD after reflecting off a diffraction grating. The 0-order reflection of the grating provides an image for alignment, and the grating can be tilted to distribute the 1st-order reflection across the horizontal CCD pixels for doing spectroscopy.



FIGURE 3.11. Put pic of new ones? Otherwise w/ asphere at least.

When a 1" diameter filter is used, the system has a collection efficiency (ϵ_c) of about 1.5% without the spectrometer, and about half of that with the spectrometer due to a grating efficiency of 50%. The spectrometer also limits the system to an f-number of 4.

To limit laser exposure to only the time of CCD exposure, a laser shutter is linked to the camera exposure with a LabVIEW program. This program also records laser power via a calibrated pickoff, as well as the temperature of the coldfinger near the sapphire window during observation.

3.6. LASER SCANNING

In order to obtain images of separated single atoms, the laser focusing lens is attached to motorized Newport translation stages which scan the laser position by scanning the lens in x and y, shown in Fig. 3.11. These stages sit atop a manual z-translation stage for laser focusing. An extension of the aforementioned LabVIEW program coordinates movement of these stages such that x or y position is stepped in between CCD frames, and each frame then corresponds to a position in a laser scan grid.

CHAPTER 4

RESULTS

Studies of several emission peaks of neutral Ba in SXe are discussed in 4.1. Temperature and annealing dependence are discussed in 4.1.1, and bleaching of these peaks is discussed in detail in 4.1.2. Imaging of Ba fluorescence in a focused laser region is discussed in 4.3, with the ultimate achievement of imaging at the single atom level using the 619-nm fluorescence peak. Candidate fluorescence peaks of Ba⁺ in SXe are reported in 4.5.

4.1. FLUORESCENCE OF BA IN SXE

Deposits of Ba in SXe absorb primarily between 540 nm and 570 nm. An absorption spectrum, obtained by observing absorption of white light by a large Ba deposit at 11 K, is shown in Fig. 4.1, along with an example emission spectrum of a Ba⁺ deposit made at 45 K and observed at 11 K with 557 nm excitation. Significant broadening, as well as a 4-nm redshift of the central peak, occur relative to the vacuum $6s^2 \ ^1S_0 \rightarrow 6s6p \ ^1P_1$ absorption value of 553.5 nm. Initial discovery of this absorption and emission was done with the purely neutral Ba getter source. Observation of the same spectra from Ba⁺ ion beam deposits demonstrates some neutralization of the ions. The fraction of ions neutralized is not determined. [18, 22, 23]

The several red-shifted emission peaks observed are attributed to Ba atoms occupying different matrix sites in the SXe. Deposition at temperatures higher than 11 K and exploration of more excitation wavelengths have led to discovery of a few emission peaks beyond the 591- and 577-nm peaks reported in [22] and [23]. Emission spectra for selected excitation wavelengths are shown in Fig. 4.2(a), for a deposit made at 44 K and observed at

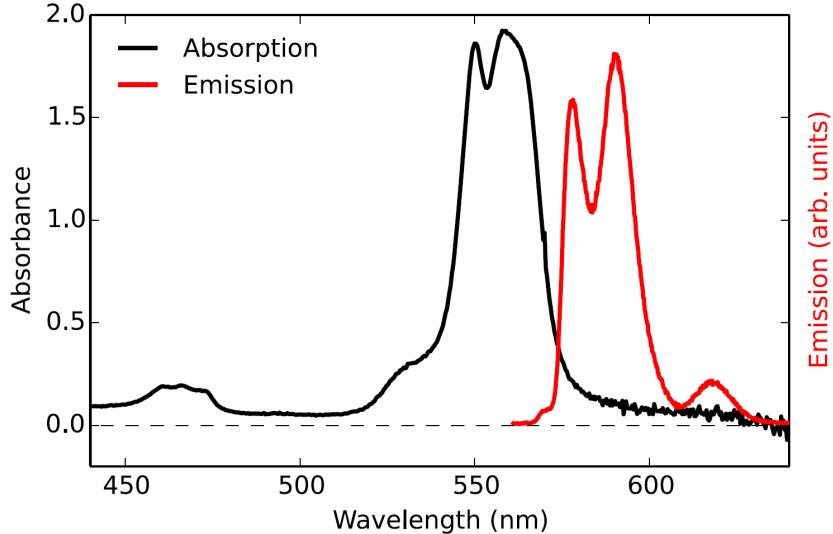


FIGURE 4.1. Absorption and emission spectra of neutral Ba in SXe. The absorption is of a Ba getter deposit at 10 K, and the emission is of a (neutralized) Ba^+ deposit, deposited at 45 K and observed at 11 K with 557 nm excitation.[18]

11 K. These selections are part of a full excitation spectrum, shown in Fig. 4.2(c). An excitation spectrum is produced by scanning the dye laser and measuring the magnitude of each fluorescence peak vs. excitation wavelength. For each 1-s CCD exposure, a sum of asymmetric Gaussian functions and standard Gaussian functions is fit to the spectrum after pedestal subtraction. The asymmetric Gaussian fit function used, which fits the 577- and 591-nm peaks somewhat better than standard Gaussians, is $A(1+\text{erf}(\frac{x-a}{\sigma_1})(1-\text{erf}(\frac{x-a}{\sigma_2}))$, where A is the free amplitude parameter. The peak position a is fixed for each peak, as are the left and right smearing widths σ_1 and σ_2 . The function $\text{erf}()$ is a Gaussian error function. The asymmetric Gaussian function is not perfect for all frames, as the shape of the emission can depend on the excitation wavelength. An example frame with fits is shown in Fig. 4.2(b). The full fit is the dotted black line, and each peak's contribution is in color. Curves in (a) have about $100\times$ the laser power used in the excitation spectrum (e.g. (b)) where low intensity is desired to avoid bleaching during the scan. Rather than attempting

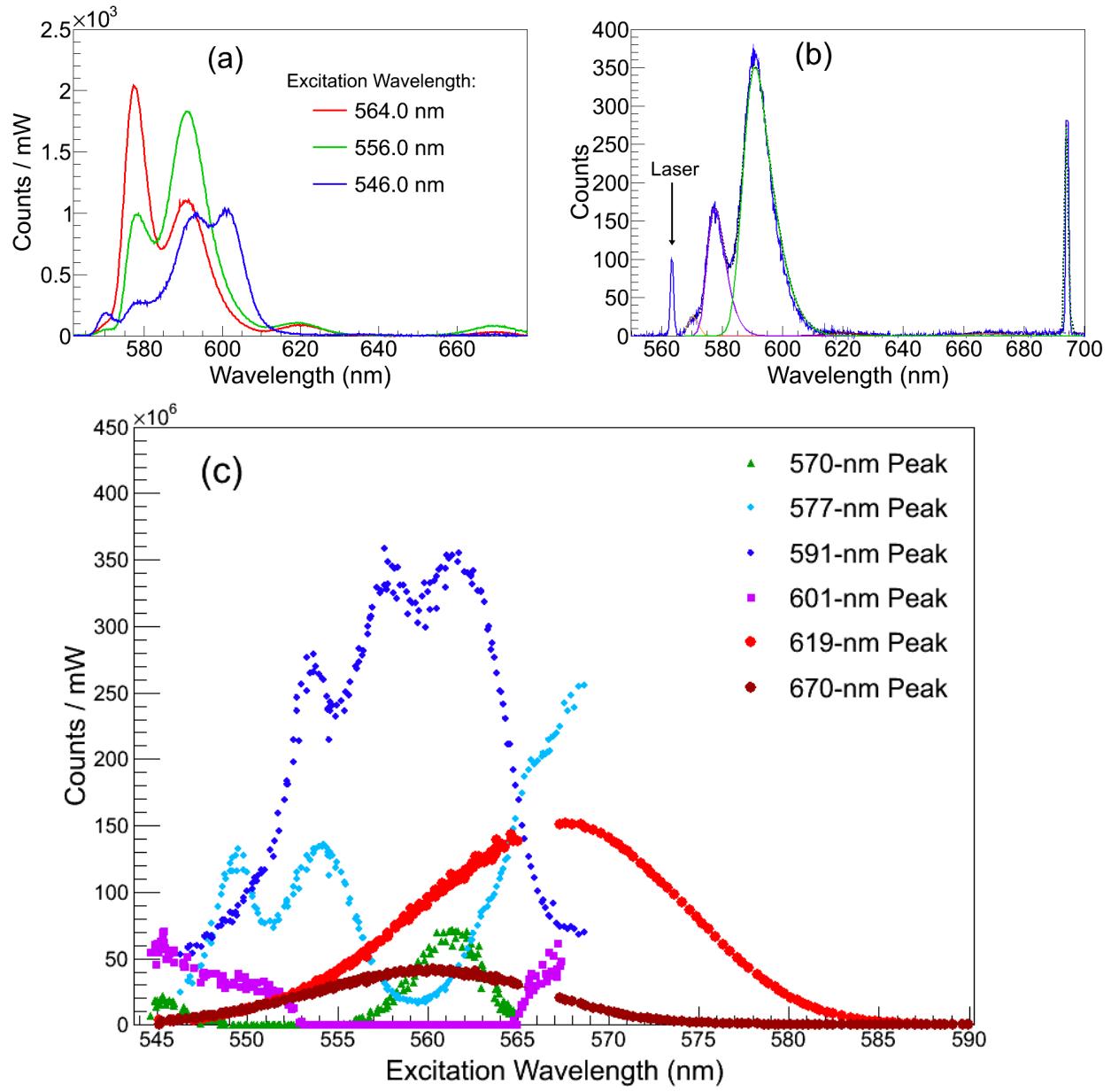


FIGURE 4.2. Background-subtracted fluorescence spectra for a few different excitation wavelengths (a), an example fit to a spectrum (b), and full excitation spectra for all observed Ba fluorescence peaks (c). Magnitudes in (c) have been scaled for visibility on the same plot (relative magnitudes are arbitrary as they are affected by relative site populations and fluorescence efficiencies). The discontinuity around 566 nm for the 619- and 670-nm peaks is the boundary between usage of different laser dyes. R6G dye is used for higher wavelengths and R110 for lower wavelengths and for all other curves. Curves for the R6G segments (619- and 670-nm peaks) require special scaling to line up with their respective R110 segments, and this scaling was different between the 619- and 670-nm peaks, likely due to different relative populations of those sites on the different deposits.

frame-by-frame background subtractions, additional Gaussians are fit to the broad and sharp background fluorescence. These backgrounds and their excitation spectra are discussed in 4.2. A 566 nm Raman filter was used to attenuate the majority of the laser scatter, however the small amount of scatter passed by the filter was used to determine the frame's excitation wavelength. Each peak's fit contribution was then integrated and scaled by the frame's laser power, as the power output is not constant through the dye range.

Wavelength calibration was done using three lasers whose wavelengths were first measured with a Burleigh Wavemeter: a red diode laser at 656.99 nm, a doubled Nd:YAG laser at 532.23 nm, and the C480 blue dye laser typically around 475 nm (at 488.91 nm for the data in Fig. 4.2). These lasers were directed at the same position on the sapphire window, and their scatter was imaged along the same path as the Ba fluorescence. The WinSpec software applies the diffraction grating equation to calibrate each CCD pixel to a wavelength.

4.1.1. ANNEALING/TEMPERATURE DEPENDENCE. Matrix site occupancies for Ba atoms can depend on annealing history, and similarly on the temperature at which a deposit is made. Spectra under different temperature conditions are shown in Fig. 4.3. Peak shapes look similar for deposits made at 40-55 K as those made at 11 K and then annealed to 40-55 K, both being observed at 11 K, though relative amplitudes are not the same, likely due to matrix site populations. Without annealing, 11-K deposits have a different peak shape around 590 nm, with a broader peak centered around 596 nm. It is possible that the broader shape is due to a higher population of the 601-nm peak, and it is not resolved from the 591-nm peak. The experiment in Fig. 4.3 did not have optimal laser intensity for observing behavior of the 619-nm peak; a study of deposit conditions for this peak is shown in Fig. 4.4. In that experiment, the 619-nm fluorescence was observed in a focused laser beam at

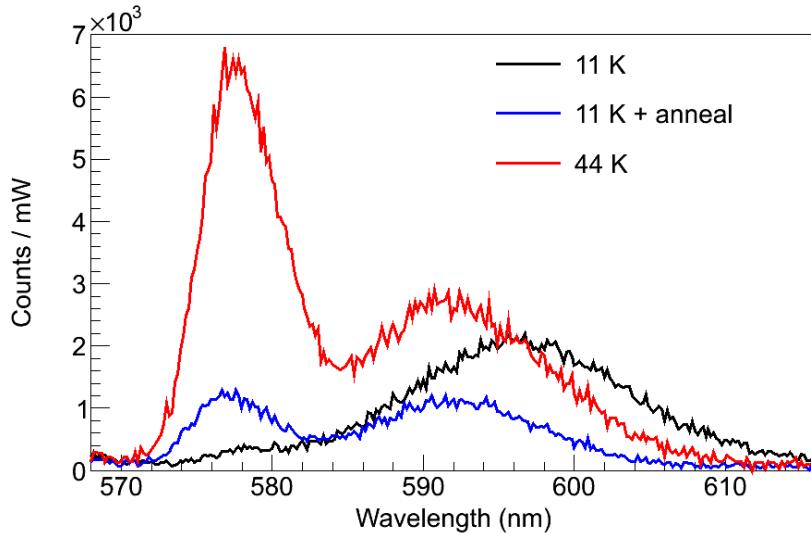


FIGURE 4.3. Spectra of peaks around 590 nm of a Ba^+ deposit made at 11 K before and after annealing to 39.4 K, and one made at 44 K. All observations are at 11 K. Both Ba^+ deposits are 15 s, however the 44 K deposit is scaled slightly to account for different ion current. Laser power was about 0.1 mW with an unfocused beam waist of $w = 7.056$ mm, at 566 nm wavelength.

570 nm, in an image through the 620-nm band-pass filter. About $3\times$ more 619-nm fluorescence was observed in the deposit at 52 K vs. 11 K for the same leak rate (the respective 31 nm/s and 37 nm/s are from the same Xe leak rate, resulting in different SXe deposition rates according to Fig. 3.9). Also in this figure is a deposit at 11 K with the lower leak rate resulting in about 5 nm/s SXe deposition, which produced nearly $3\times$ lower signal than the leak rate resulting in 37 nm/s SXe deposition. Using the 10 K SXe density of 3.780 g/cm^3 [26] and a typical Ba^+ ion current density of 1.6 nA/mm^2 at the sapphire window, 5 nm/s and 37 nm/s correspond to Xe:Ba ratios of about 8.7×10^3 and 6.4×10^4 respectively. Xe leak rates above 37 nm/s result in rapid frosting of the SXe matrix, which causes blurring of the image and high laser scatter. These tests guided the standard of depositing with Xe leak rate resulting in 31 nm/s SXe deposition at 50 ± 5 K when observing the 577-, 591-, and/or 619-nm peaks. The different bleaching behavior in the low Xe leak rate deposit is interesting but not well understood.

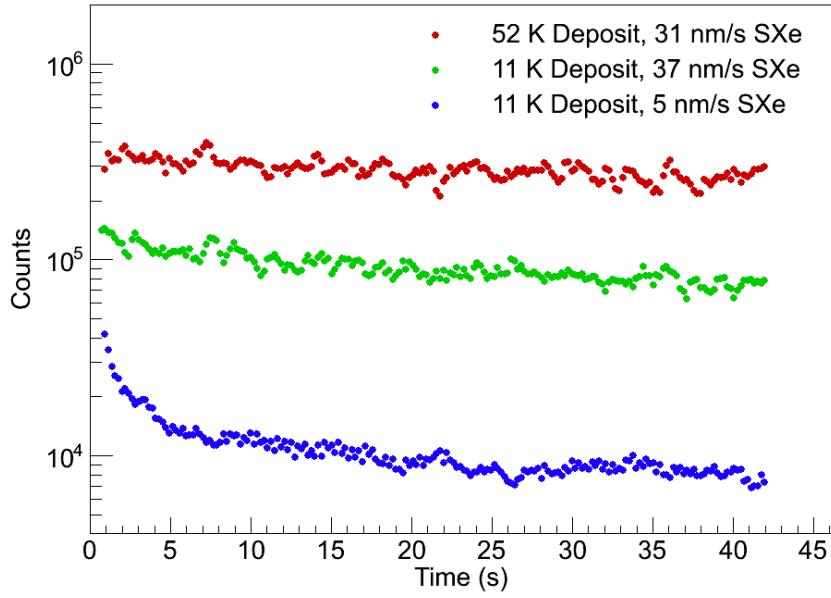


FIGURE 4.4. 619-nm fluorescence counts over time from imaging focused 570-nm laser region, for Ba^+ deposits at different temperature and Xe leak conditions. Frame times are 0.1 s, with time in between frames of about 0.11 s.

Fluorescence spectra with 564 nm excitation through several annealing cycles for a deposit made at 11 K are shown in Fig. 4.5. At this wavelength, all observed Ba peaks are prominent except the 570-nm peak. Similar to excitation spectra, individual fluorescence peak amplitudes can be plotted vs. temperature by fitting the full spectrum in each frame. Rather than asymmetric Gaussians, this analysis used standard Gaussian and Lorentzian functions. Lorentzians fit the 619-nm and 670-nm peaks well. Each individual peak's amplitude is plotted vs. temperature in Fig. 4.6(b-f), with an example of a fit spectrum in Fig. 4.6(a). In this interpretation, the 601-nm peak along with the 591-nm peak fit the broader peak in the initial 11 K deposit. The 601-nm peak had nearly complete loss in the first anneal cycle. The 670-nm peak had significant loss, with more loss after each succeeding anneal cycle, each of which reached higher a temperature than the last. The 591-nm had moderate loss with each cycle. The 577-nm and 619-nm peaks gained significantly with the first anneal, suggesting that they are due to more stable matrix sites. Both peaks remained

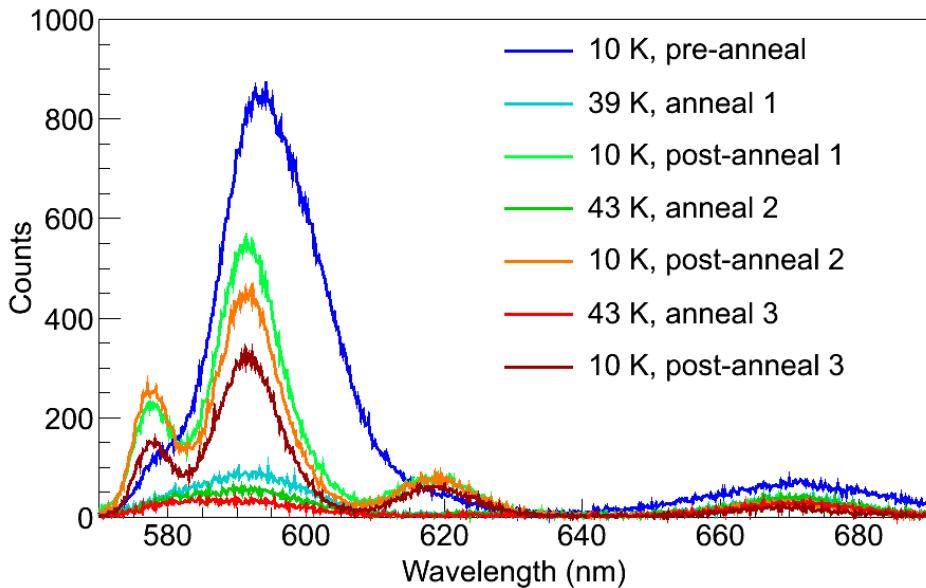


FIGURE 4.5. Spectra of a large Ba^+ deposit through several annealing cycles. Initial deposit was at 11 K. Laser power was about 0.2 mW with an unfocused beam waist of $w = 7.056$ mm, at 564 nm wavelength. These are selections from the full data set shown as peak counts vs. temperature in Fig. 4.6. *This same data, but a different plot, is in BaSpec, as is some of the annealing plots – do I just cite the paper or what? rm things like that and write them in blue w/ pen on printout*

about the same after the second cycle (small gain in 577-nm), and both had loss after the third cycle, which reached the higher temperature of 48 K. This could be due to greater diffusion at higher temperature leading to Ba/Ba interactions.

Aside from matrix site changes, direct temperature dependence of fluorescence can be observed in annealing cycles. The 577-, 591- and 619-nm peaks have their highest amplitude at 11 K. The 577-nm and 619-nm peaks appear to be leveled off at 11 K, while the 591-nm may benefit from even lower temperatures. The 670-nm peak has its highest amplitude at around 25 K (apparent after its major site alteration has occurred). This inverse relationship between fluorescence and temperature suggests that a probe in nEXO will need to be moved to an evacuated chamber in order to cool to 11 K or below for observation.

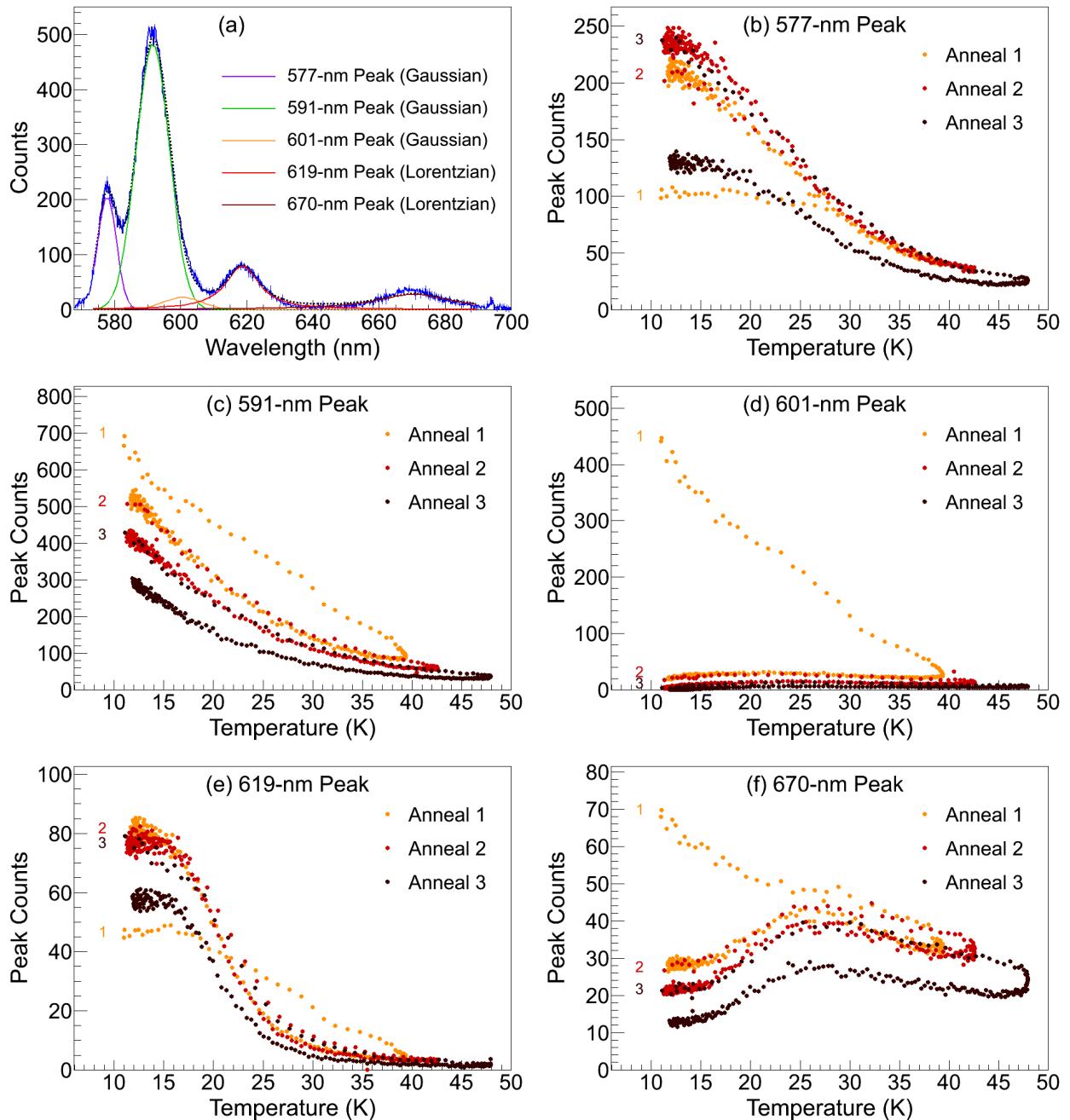


FIGURE 4.6. Example fit with standard Gaussians and Lorentzians (a) and fit peak counts for each fluorescence peak through three annealing cycles of the Ba^+ deposit made at 11 K (b-f). “1, 2, and 3” mark the beginning of each anneal cycle. Laser power was about 0.2 mW with an unfocused beam waist of $w = 7.056$ mm, at 564 nm wavelength.

4.1.2. BLEACHING. Decay of fluorescence with laser exposure, or bleaching, was observed for all six Ba fluorescence peaks. It is a rapid process for the 570-, 577-, 591-, and 601-nm

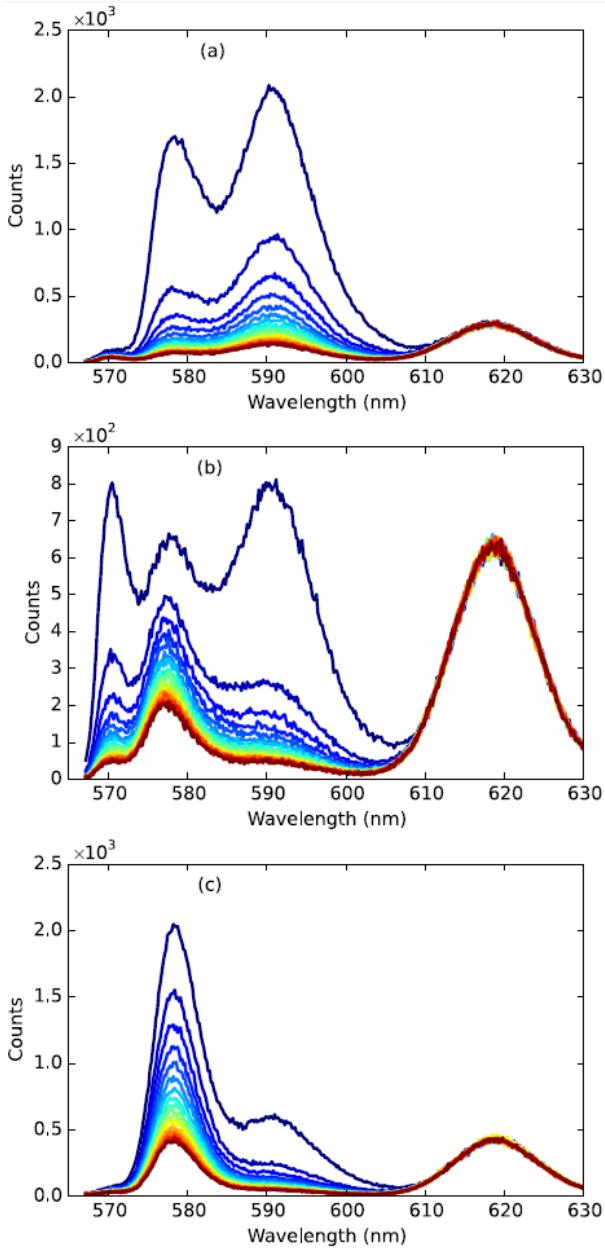


FIGURE 4.7. will re-do these, ideally showing 670-nm. [18]

peaks, and a much slower process for the 619- and 670-nm peaks. With the prospect of counteracting the effect for single-atom imaging, bleaching of the 577- and 591-nm peaks was studied in detail by observing the fluorescence decay rate for varying excitation rates.

To produce a well-known laser intensity, the laser was de-focused to a specific beam waist (w), and the image of 619-nm fluorescence (which defines the laser region due to its

low bleaching) from a large Ba⁺ deposit was centered in the spectrometer slit and a y-pixel region of interest (ROI) with edges at 90% of the maximal intensity. Ba spectra over time are shown in Fig. 4.7(a,b,c) for excitation at (a) 556.9 nm, (b) 562.6 nm, and (c) 566.3 nm, where relative bleaching rates between peaks can be seen to depend on the excitation wavelength. Fluorescence counts vs. total excitations (excitation rate $w_{12} \times$ time) for the 577- and 591-nm peaks are plotted in Fig. [fig bleaching]. ...I swear this doesn't make sense, because you KNOW $1/x \times t$ and $x \times I$ does not yield the same total counts. **don't forget the correction on p-meter sensitive area and on p-meter quantum efficiency**

The 6-level model solved in [sec. in Ch. 2] can be applied to this bleaching data to determine deviations from vacuum transition rates. The 6th level allows for one additional vacuum-forbidden transition from the excited state, a_{26} and its decay back to ground a_{61} , e.g. a ³P state. A successful model will fit observed bleaching curves for all values of w_{12} with the same transition lifetimes. An example *examples of* of a good model fits is shown in Fig. 4.8. The nearly constant decay slope in later frames was easily fit with the introduction of non-zero a_{26} , and could not be fit by manipulation of the existing transitions, suggesting the presence of an additional matrix-induced path out of the excited state. In addition, non-zero a_{61} values only hurt the fit, suggesting that this state does not have a spontaneous recovery path (*is this just for one of the peaks?*). An important observation is that the other rates do not require much change from their respective vacuum values, with the exception of *whatever that one is. try non-changed $a_{2?}$, to see if maybe the extra .5-s is actually in between each frame ... go into the lab and try come where this ,5-s would dominate and see if it's there* This suggests that the observed bleaching is mostly due to optical pumping into

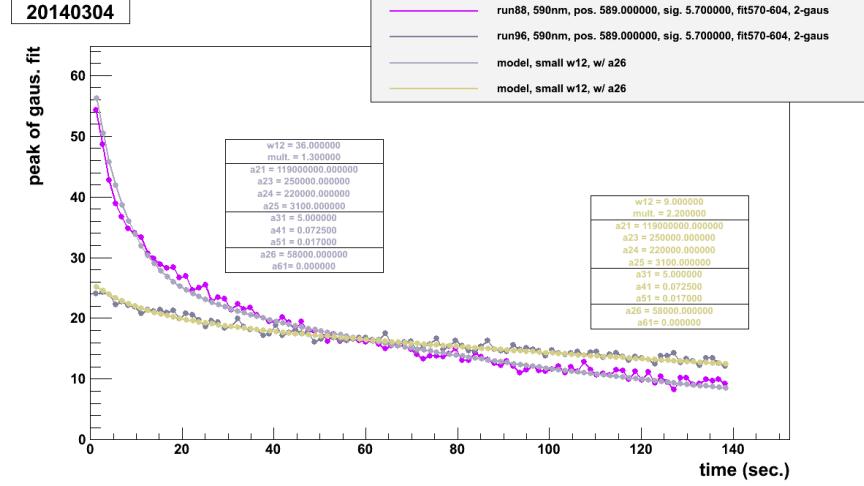


FIGURE 4.8. example place-holder

the metastable D states, with the exception of a_{26} , and thus that laser re-pumping should be possible to increase the fluorescence signal from these peaks.

Another interesting result is the model's inability to explain the low fluorescence efficiency (ϵ_f) observed in these peaks. The number of counts observed is significantly lower than expected from $a_{21} = ?$. ϵ_f , defined in [Ch.2], was measured by the total counts observed in the first frame of exposure divided by its respective w_{12} , shown in Fig. 4.9. This method of measuring ϵ_f becomes less accurate with more optical pumping in the first frame, i.e. with higher w_{12} for a given frame time. Thus, the trend should be extrapolated toward zero, suggesting a real ϵ_f of about 1%. The bleaching model was unable to explain this low ϵ_f by additional paths out of the excited state without introducing extreme alterations to existing rates, or by introducing extreme a_{26} and a_{61} , in order to dominate a_{21} . A possibility is that the low count rate actually reflects a matrix site population by only about 1% of deposited Ba⁺ ions.

Effecting re-pumping of optically pumped Ba could require up to three additional lasers, one for each of the populated metastable D states, as it does in vacuum. The optimal

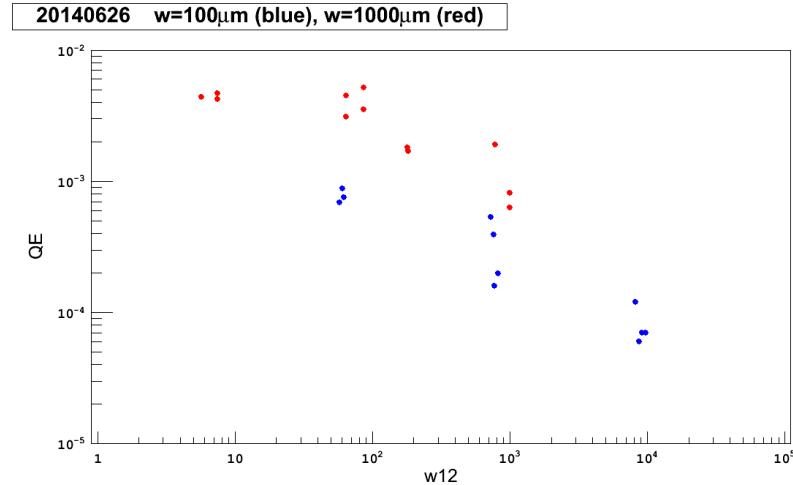


FIGURE 4.9. *correct this too for real w12*, and needs some correction to combine the two regions?

absorption for each transition would need to be discovered using tunable lasers in the infrared, and the search would be challenging, as the effect of re-pump is small until all three are achieved. However, mixing of states in the Xe matrix could lower the number of lasers needed. Searches for an effect of additional excitation lasers were attempted with a few different lasers. *describe which lasers were supposed to do which transitions – 1550, 1064, 657, 406, blue bye.*

The only effects observed on bleaching rates by these re-pump lasers were increased bleaching by the blue dye and 406-nm Kr ion lasers. Some effect of a return of fluorescence was observed after separate exposures of low-intensity 406-nm Kr ion laser, shown in Fig. [fig Kr return]. In this experiment, bleaching of the 591-nm peak was observed by x-nm excitation, and then the x-nm laser was blocked for a length of time, either for a waiting period or for exposure to the 406-nm laser. Larger returns in 591-nm fluorescence were observed with 406-nm exposure than for periods of just waiting, for low intensities of the 406 nm. However, co-exposure of this intensity of 406 nm with the green excitation laser had no noticeable effect. This phenomenon was not explored further.

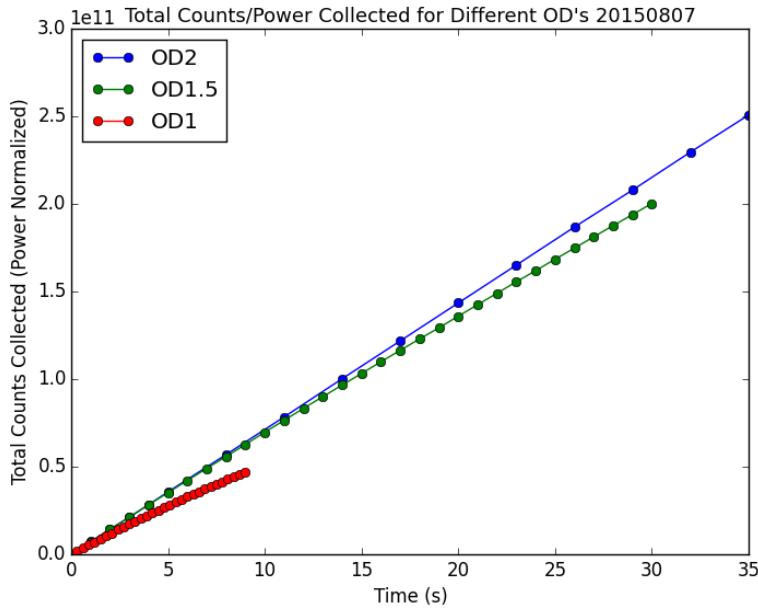


FIGURE 4.10. needs Tim plot

The 619-nm peak bleaches at much higher laser intensities. At a few mW of around 570 nm, bleaching is observed when the laser is focused. Integrated 619-nm fluorescence, divided by laser power, vs. time from an image of a focused 570-nm laser is shown in Fig. 4.10 for three laser powers. Observation of lower total counts when laser intensity and exposure time are increased and decreased, respectively, by the same factor, i.e. the same number of photons on faster time scales, indicates a time-dependent return mechanism for the fluorescence. This could be a return from a metastable state. Alternatively, if the 619-nm bleaching is caused by laser-heating of the sample (likely by heating the sapphire window), then the time-dependent return could be defined by the cryostat cooling power. Faster exposure times are more convenient, however this must be balanced against a loss of fluorescence. The x mW/ μm^2 intensity was chosen, with exposure times of around 60 s.

4.2. BACKGROUNDS

One source of background emission was observed from the surfaces of the window. Its broad fluorescence is shown in Fig. 4.11(a) with a 610-nm Raman filter cutoff and 570.4 nm excitation, and its excitation spectrum is shown in Fig. 4.11(b) over the R6G dye range. The nature of this emission has not been determined, however a few features were identified. One was that the emission increased as the window temperature was decreased, down to about 100 K where it remained flat down to 11 K, shown in Fig. 4.12. Another feature of the surface background is that it bleaches with laser exposure. This was useful in that the background could be reduced by pre-bleaching of the window. However, the bleaching behavior also presented an inconvenient time dependence of the surface background, as historical bleaching and possible slight movements of the laser could lead to variation. Frequent Xe-only deposits were made in order to establish proper background subtraction. Spacial variation in the background is especially cumbersome in a laser scanning experiment, as discussed in 4.4. Given these behaviors, it is possible that the surface background is caused by a species which freezes to the window, or something which coats the window and fluoresces more at lower temperatures. In either case, the bleaching could be explained by evaporation with laser heating, or by optical pumping of the species into a metastable state.

Another source of background is fluorescence from the sapphire window. A spectrum of this fluorescence with 562 nm excitation is shown in Fig. 4.13(a). The strong, sharp peak at 694 nm is a well-known emission of Cr³⁺ impurities in the sapphire bulk. An excitation spectrum for this peak is shown in Fig. 4.13(b) over the range all three dyes R6G, R110, and C480, using a sapphire window with relatively high Cr³⁺ content. Multiple features observed in the excitation spectrum are consistent with the absorption spectrum of Cr³⁺ in

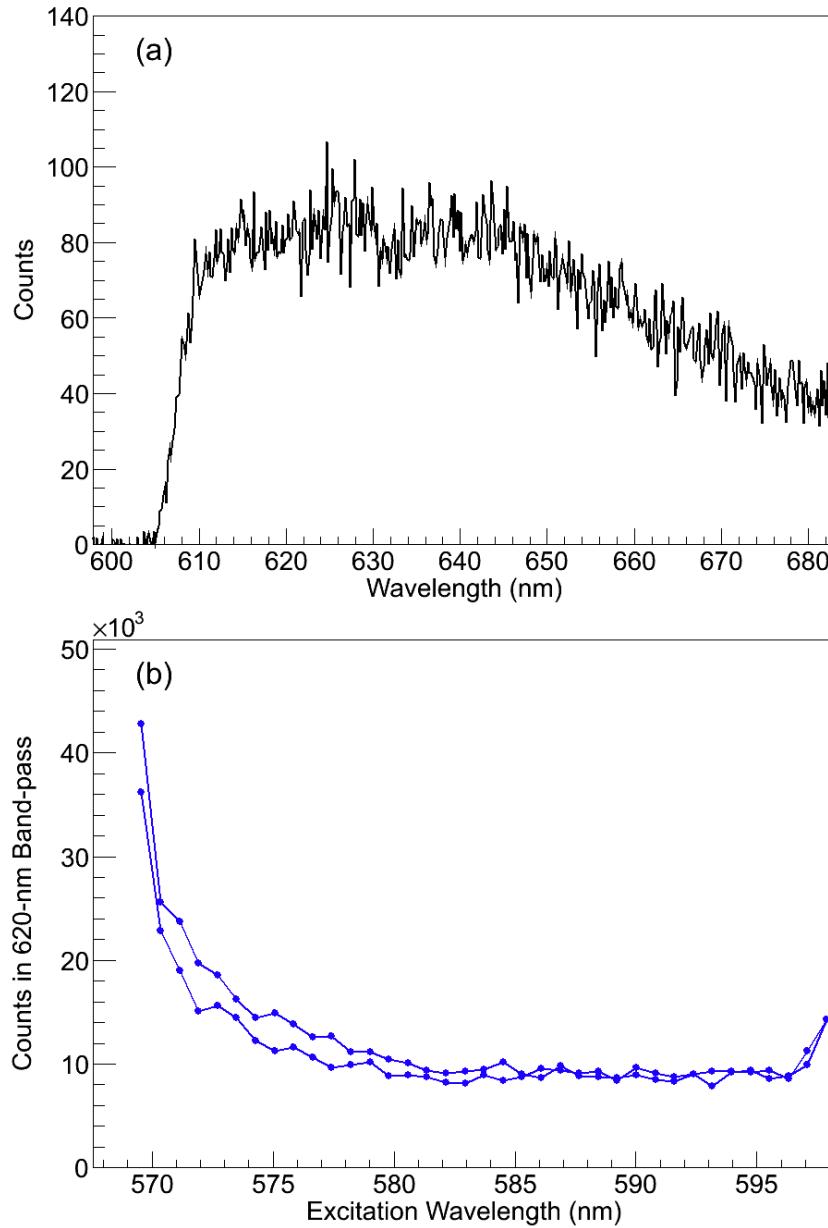


FIGURE 4.11. Surface background emission spectrum w/ excitation at 570.5 nm (a) and its excitation spectrum in R6G dye range (b). The sharp drop in (a) around 608 nm is the Raman filter cutoff.

sapphire at 77 K, including three sharp peaks in the blue, and a broad absorption in the green/yellow with vibrational peaks on the red tail [27, 28]. In addition to the 694-nm peak, a weaker and much broader emission is observed, along with three weak peaks around the 619-nm Ba peak region, also from the sapphire bulk. Excitation spectra for these fluorescence

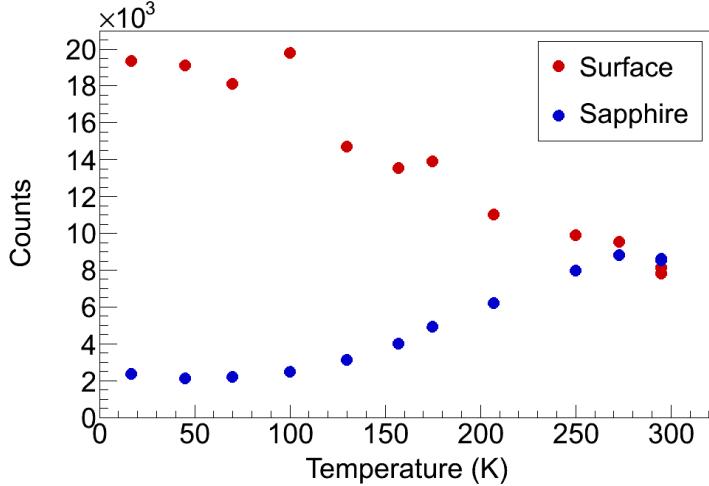


FIGURE 4.12. Temperature dependence of surface (red) and sapphire bulk (b) backgrounds.

components are shown in Fig. 4.14 for the R6G dye range. In this experiment, the laser was de-focused to about $w = 200 \mu\text{m}$, and the emission observed was from the surface laser spot. As a result, there is contribution of the surface background in the spectrum; this is negligible for the prominent 694-nm peak, however its features, broad excitation with rises near 600 nm and 567 nm, can be seen in the excitation spectra of the weaker components (blue, and especially orange curves in Fig. 4.14). Nonetheless, observation of the same vibrational peaks as in the 694-nm peak demonstrates that the broad emission and weak peaks in the 620 band-pass (Fig. 4.13(a)) are also due to Cr^{3+} in the sapphire. Commercially available c-plane quality sapphire windows contain very low concentrations of Cr^{3+} . from a few companies were tested, and those from Meller Optics produced the lowest sapphire bulk emission in the 620-nm band-pass region. The choice of 570 nm for excitation of the 619-nm fluorescence peak was made by an optimization of S/\sqrt{B} , where S is the 619-nm signal and the background B is the broad emission in the 620 band-pass filter region (orange curve in Fig. 4.14). The Cr^{3+} emission has an inverse relationship with temperature, shown in Fig. 4.12.

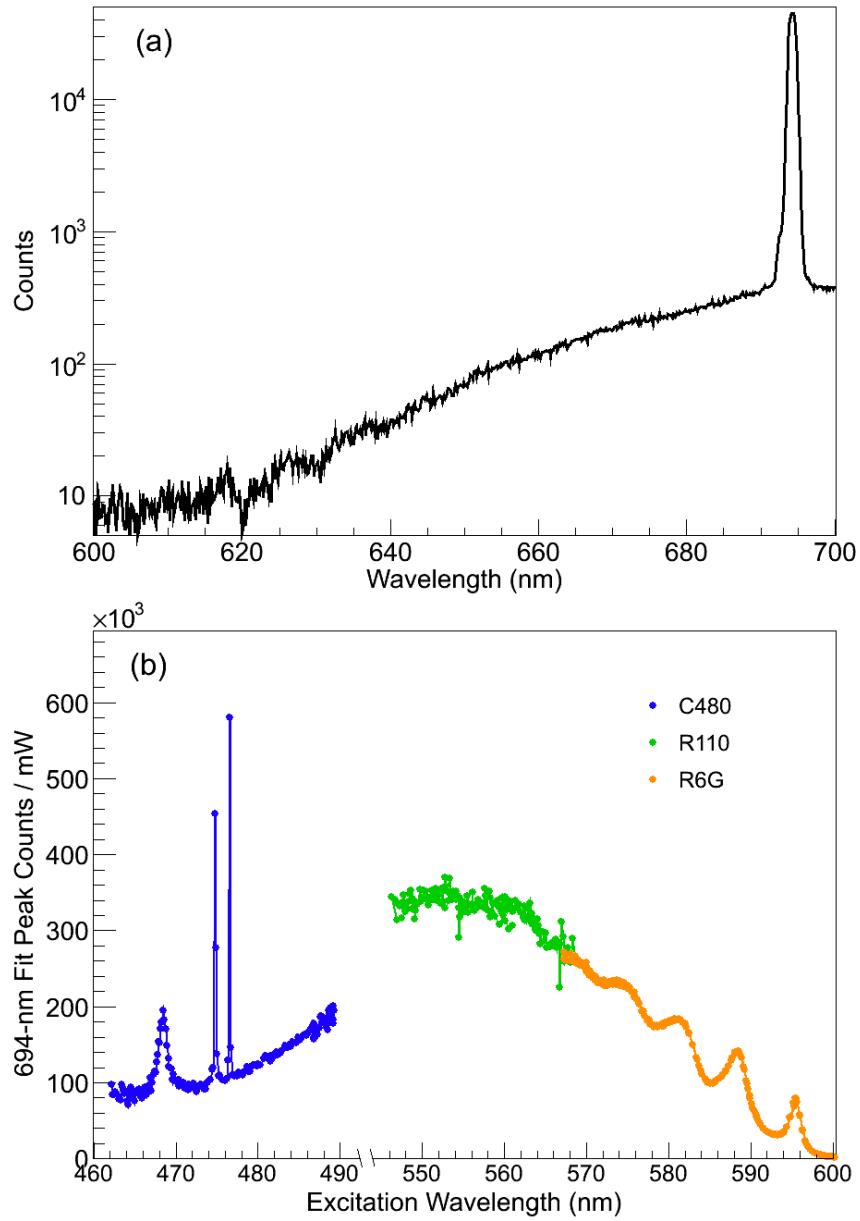


FIGURE 4.13. Sapphire bulk emission with 562-nm excitation at 11 K (a), and excitation spectrum of the sharp 694-nm emission peak using three different laser dyes (b). Relative heights of Coumarin and Rhodamine are not exact, as excitation rates are different between experiments. R110 and R6G were scaled to match at their boundary for the same reason.

4.3. IMAGING

Though the imaging spectrometer can produce spacial images with the 0-order grating reflection, better collection efficiency and imaging quality are achieved by removing the

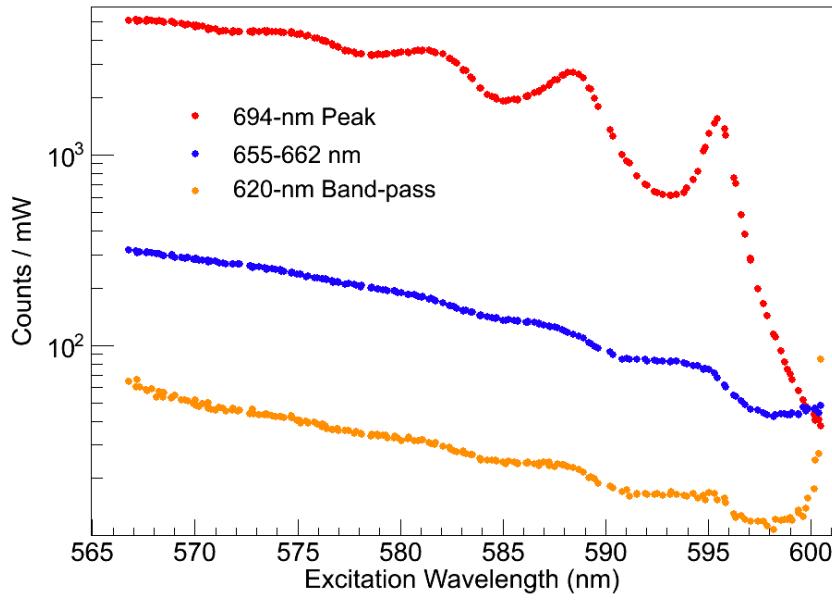


FIGURE 4.14. Excitation spectra for weaker sapphire bulk emissions (blue,orange) along with that of the sharp 694-nm emission (red).

spectrometer and imaging directly onto the CCD. Band-pass filters were used to pass the desired Ba fluorescence peak(s) while attenuating laser scatter and sapphire fluorescence.

An example image of the focused 570 nm dye laser passing through a c-plane sapphire window of thickness 0.5 mm, using the 620-nm band-pass filter on the fluorescence, is shown in Fig. 4.15. With 4 \times magnification, each pixel is 5 $\mu\text{m} \times 5 \mu\text{m}$. The laser's path through the window is faintly visible by the tail of the broad Cr³⁺ fluorescence in the 620-nm band-pass. The laser is focused at the top surface of the window, which faces the ion beam. The surface background is seen on both surfaces. The observed size of the laser spot, with a 1/e² radius of about 12 μm , is larger than the 2.06 $\mu\text{m} \times 2.66 \mu\text{m}$ 1/e² laser spot size. Aberrations and vibrations in the collection optics could contribute to this inability to reach the diffraction limit in imaging.

4.3.1. VIBRATIONS AND EFFECTIVE LASER REGION. Relative vibrations between the laser and sapphire window will affect the number of Ba atoms exposed, increasing the effective

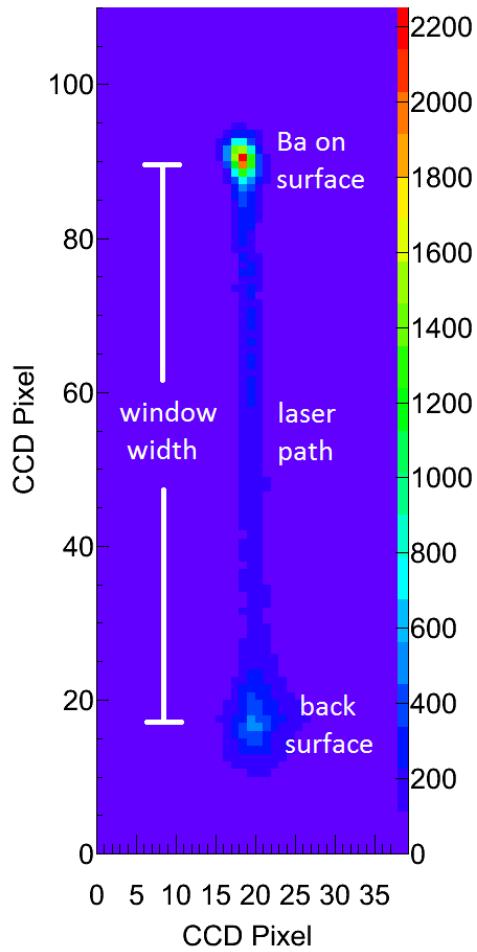


FIGURE 4.15. Example image of focused laser spot on sapphire window surface, and its path through the window. *will replace w/ one w/o Ba*

laser spot size. This was studied by observing the position of a “dust spot” (a highly scattering feature on the sapphire window) relative to the position of the laser in an image on time scales down to 50 ms. An example of an image from this experiment is shown in Fig. 4.16. The 570-nm dye laser was somewhat de-focused, and the dust spot was illuminated by a de-focused 657 nm diode laser. For each frame, 2D Gaussian functions were fit to locate the center of the laser spot and the dust spot in order to measure their relative position. The fit for the laser spot was restricted in y so that it was not affected by the bulk sapphire fluorescence path. The distances in x and y between the dust spot and laser are plotted for

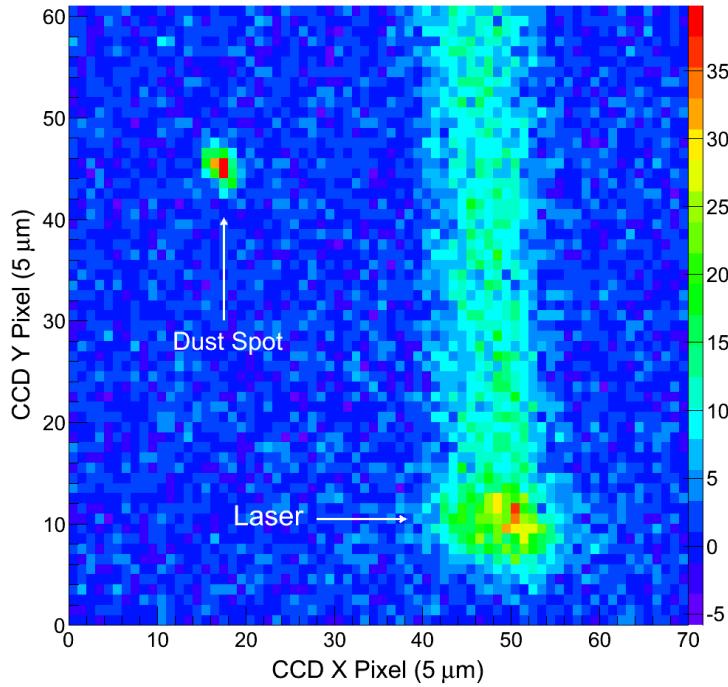


FIGURE 4.16. Example image of dust spot and laser during observation of cryostat vibrations.

each 50-ms snapshot in Fig. 4.17(a). The distribution shows a correlation between x and y. To calculate an effective laser spot size due to this vibration, each difference (x,y) between laser and dust spot was used as the center of a 2D Gaussian, each with $w_x = 2.06 \mu\text{m}$ and $w_y = 2.66 \mu\text{m}$ to represent the laser spot. Such Gaussian functions were summed to all points to produce a distribution of summed laser exposure, shown in Fig. 4.17(b). The total area enclosed by a $1/e$ contour then represents the effective area. Since x and y movement are correlated, and since the movement is sinusoidal, the effective area is only about $2 \times$ the real laser spot size, and the effective laser region is $17.8 \mu\text{m}^2$.

The aforementioned is the most concerning vibration to understand. Vibration of the laser itself is included in that study. Vibration of the collection optics will affect the imaging

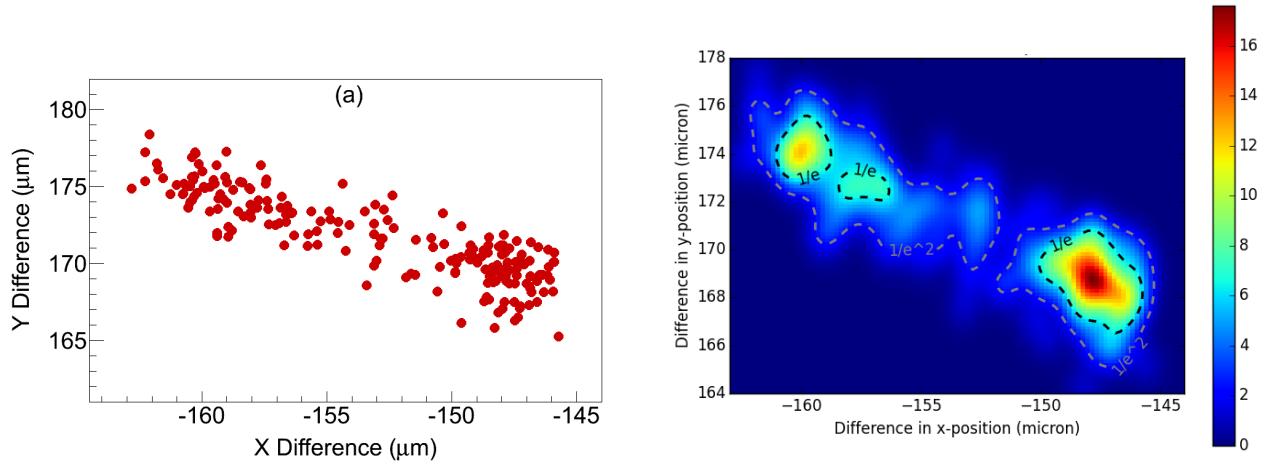


FIGURE 4.17. Cryostat vibration measurements based on relative position of dust spot vs. laser on sapphire window in 50-ms snapshots (a), with 2D Gaussians overlain on each point to represent total laser exposure vs. position (b). *will re-do (b) myself*

resolution, but not the number of atoms being observed. These vibrations were minimized by stable mounts.

4.3.2. IMAGING 577- AND 591-NM PEAKS. First attempts at imaging small numbers of Ba atoms in a focused laser region were done with the 577- and 591-nm Ba fluorescence peaks together using a 586-nm band-pass filter, which passes 573 - 599 nm FWHM. This filter has a 2" diameter, resulting in a factor of 4× more collection efficiency. Bleaching data was used to optimize laser intensity and exposure time to achieve maximal signal in frame 1, and to avoid hole bleaching. The fast bleaching of these peaks is the limiting factor on sensitivity. An image of $\leq 10^4$ atoms (10^4 Ba⁺ ions deposited into the effective laser region) is shown in Fig. 4.18. This is a 100-s exposure with 0.02 μW of 566-nm excitation. At this low intensity, no bleaching was observed in the four frames observed (frame 1 is shown). Since the effective laser region is approximately doubled by cryostat vibrations, the signal observed at a given moment in time is actually due to the real laser spot size in the scenario of little to no bleaching. Thus it should be said that Fig. 4.18 is an image of 5×10^3 atoms on average,

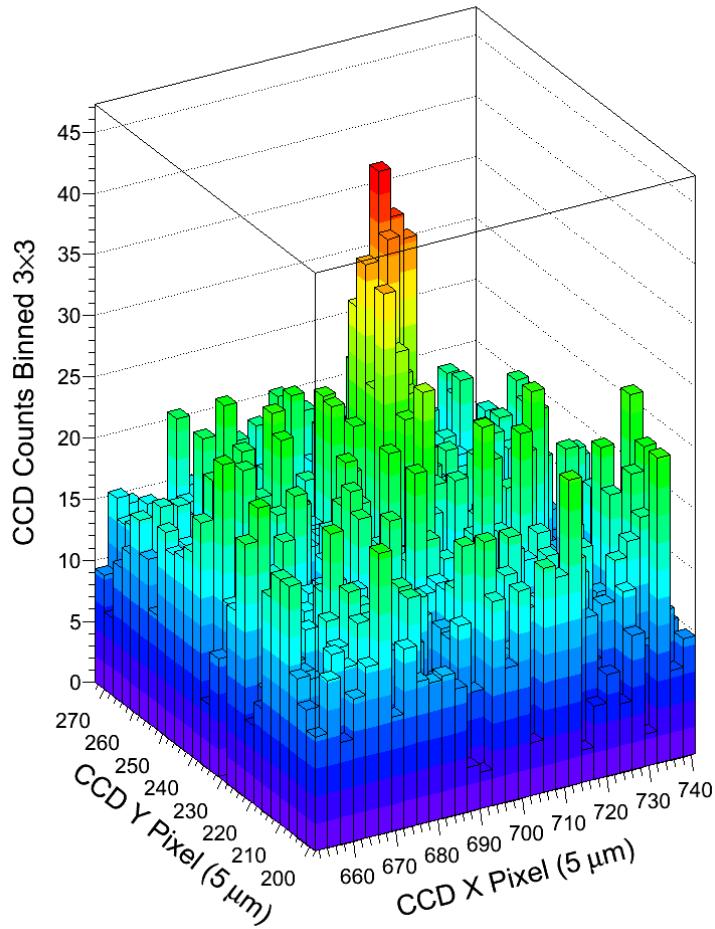


FIGURE 4.18. Image of about 10^4 total Ba atoms in SXe, with about 5×10^3 atoms exposed at a time. 100-s exposure with $.02 \mu\text{W}$ of 566 nm excitation, deposited at 50 K and observed at 11 K. 3×3 CCD pixel binning.

with 10^4 total atoms exposed. Groups of 9 (3×3) CCD pixels are binned to produce the nice peak shown. As discussed in 4.1.2, detection of very small numbers of atoms in these sites may require several infrared re-pump lasers. As a result of low total exposure, neither the sapphire nor the surface backgrounds are present in these images, though a Xe-only image has been subtracted.

4.3.3. IMAGING 619-NM PEAK. At low laser intensities, the 619-nm peak appears minor compared to the 577- and 591-nm peaks. However, its much lower bleaching rate brings it to

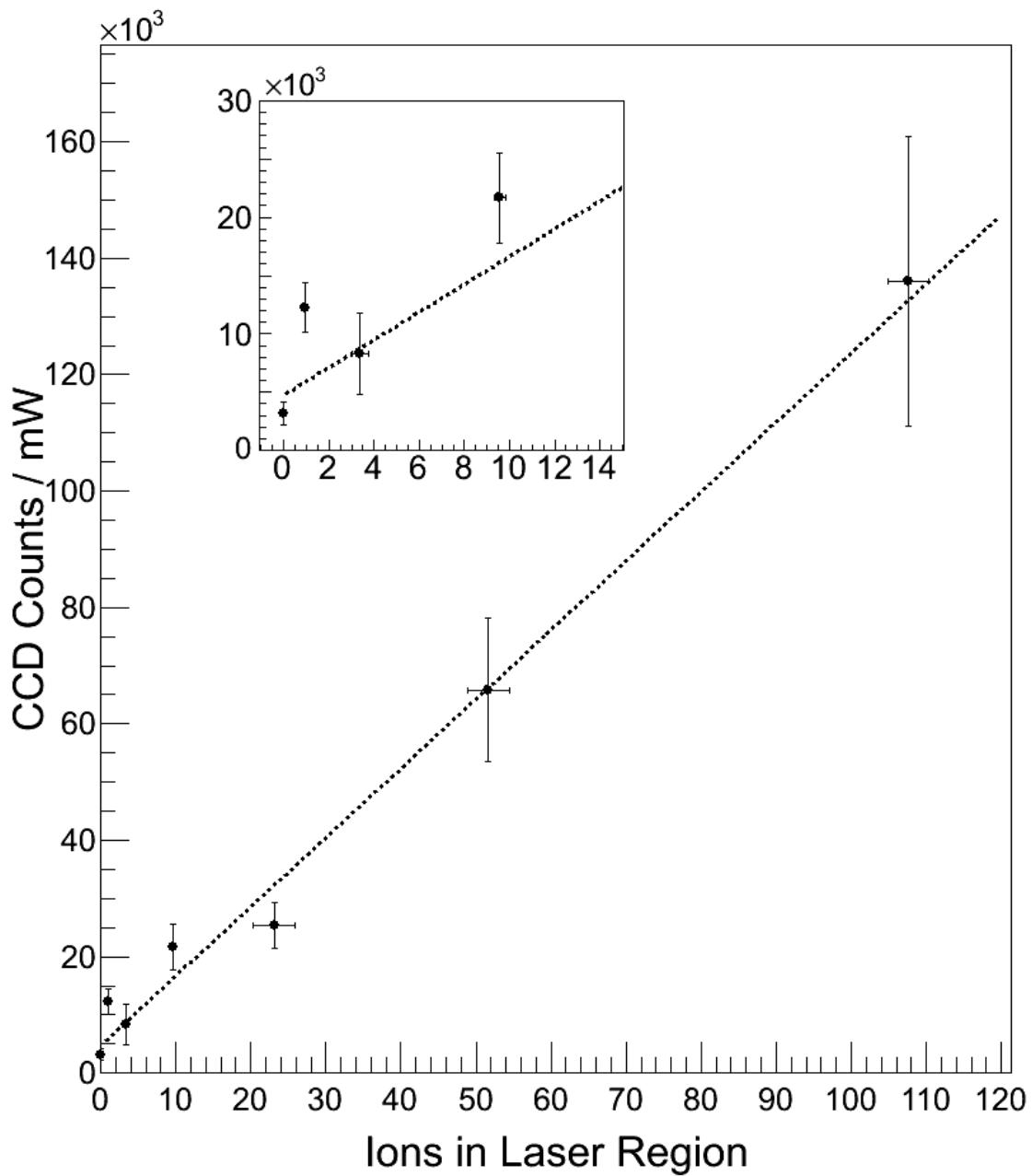


FIGURE 4.19. 619-nm signal vs. Ba^+ deposited in focused laser region. *move y-ax title over, Remove x errors*

dominate over larger exposures, e.g. with the intensity of a focused laser at 0.01-0.1 mW over seconds or minutes. Experiments imaging small numbers of Ba atoms in SXe in a focused laser region with the 619-nm peak were successful down to the single-atom level.

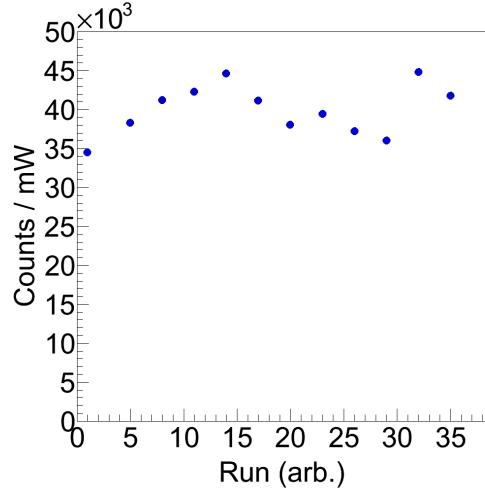


FIGURE 4.20. Background counts in focused laser region over the course of an experiment.

An imaging experiment consisted of many different pulsed Ba^+ deposits, each of which was evaporated after observation. This procedure is described in 3.3-3.5. A linear relationship between observed signal and ions deposited must be observed and repeated. Frequent Xe-only deposits were made, typically every three runs, to establish the local background. The image of each deposit was analyzed by summing the counts of a $3\text{-pixel}\times 3\text{-pixel}$ ($15\mu\text{m}\times 15\mu\text{m}$) region centered on the laser spot in the SXe layer where it excites Ba atoms, i.e. at the top of the laser path in Fig. 4.15. A partial-bin integrator was implemented such that the $3\text{-pixel}\times 3\text{-pixel}$ region could be centered on the peak center of each run, which typically varied by about half a micron between runs. Summed counts vs. Ba^+ ions deposited into the laser region is shown in Fig. 4.19 for a combination of data sets from two separate days *maybe you want them separate (with each point having errors) by color to show agreement*. Each point has subtracted the averaged counts from the two surrounding Xe-only deposits. Some variation in signal is observed, likely due to spatial drifting of the ion beam, as this variation was larger on days where larger beam drift was observed. Nonetheless, a clear linearity is observed in signal vs. ions deposited, all the way down to

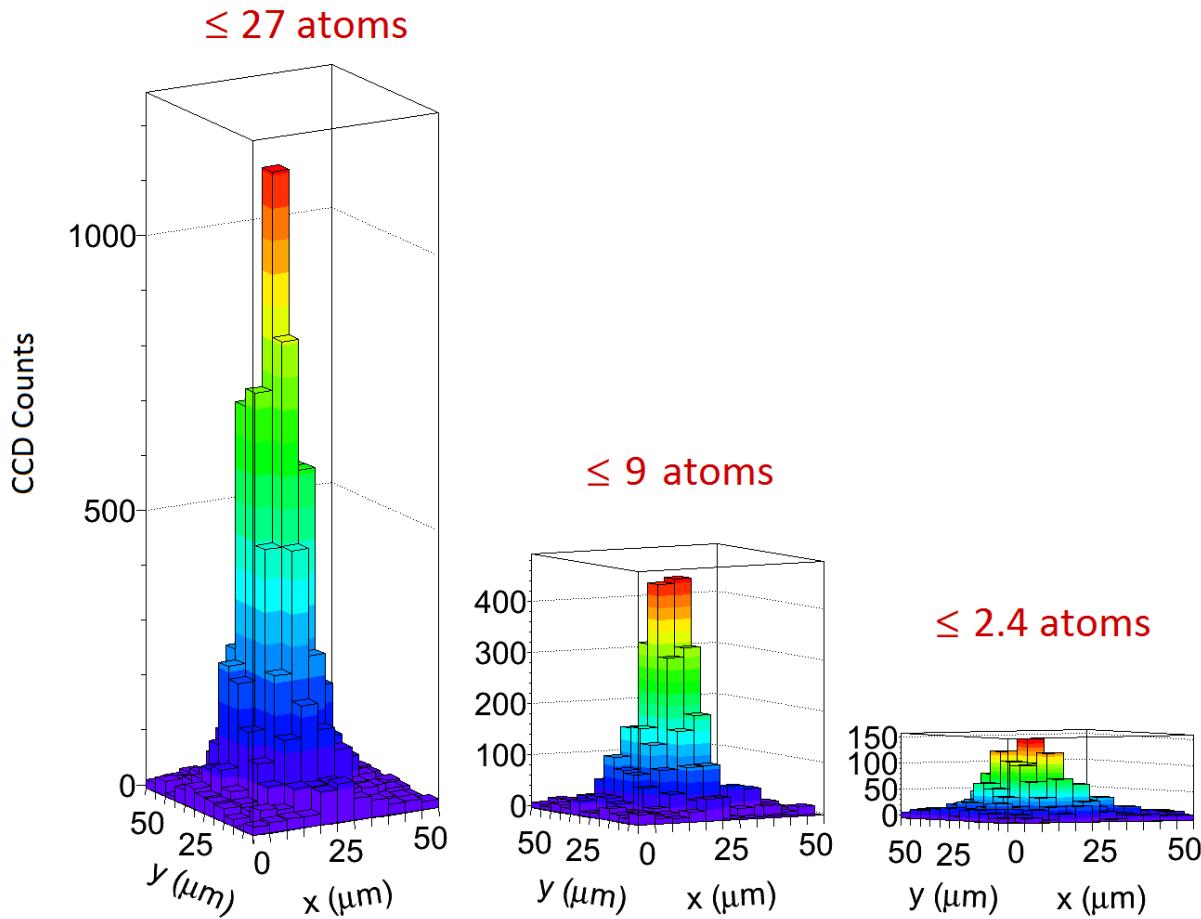


FIGURE 4.21. will change: 1) have more than this, 2) do pixel (5micron) on axis like others, 3) have same-looking phi angle on all

single ions deposited. Recall that the number of ions is an upper limit on the number of atoms in the 619-nm peak matrix site. “0-pulse” deposits are also made, wherein Faraday cup 3 is retracted for the 1-s period, but the ion beam remains deflected with no pulses. This establishes the level of neutral Ba atoms making their way down the beam line from the source into the laser region. Signal at the few-atom level is observed from these deposits.

Variation in the background level, dominated by the surface background, is shown in Fig. 4.20. Variation was most likely caused by drift of the laser position on the window, to regions of different historical bleaching. Local variations are at the single-atom signal level,

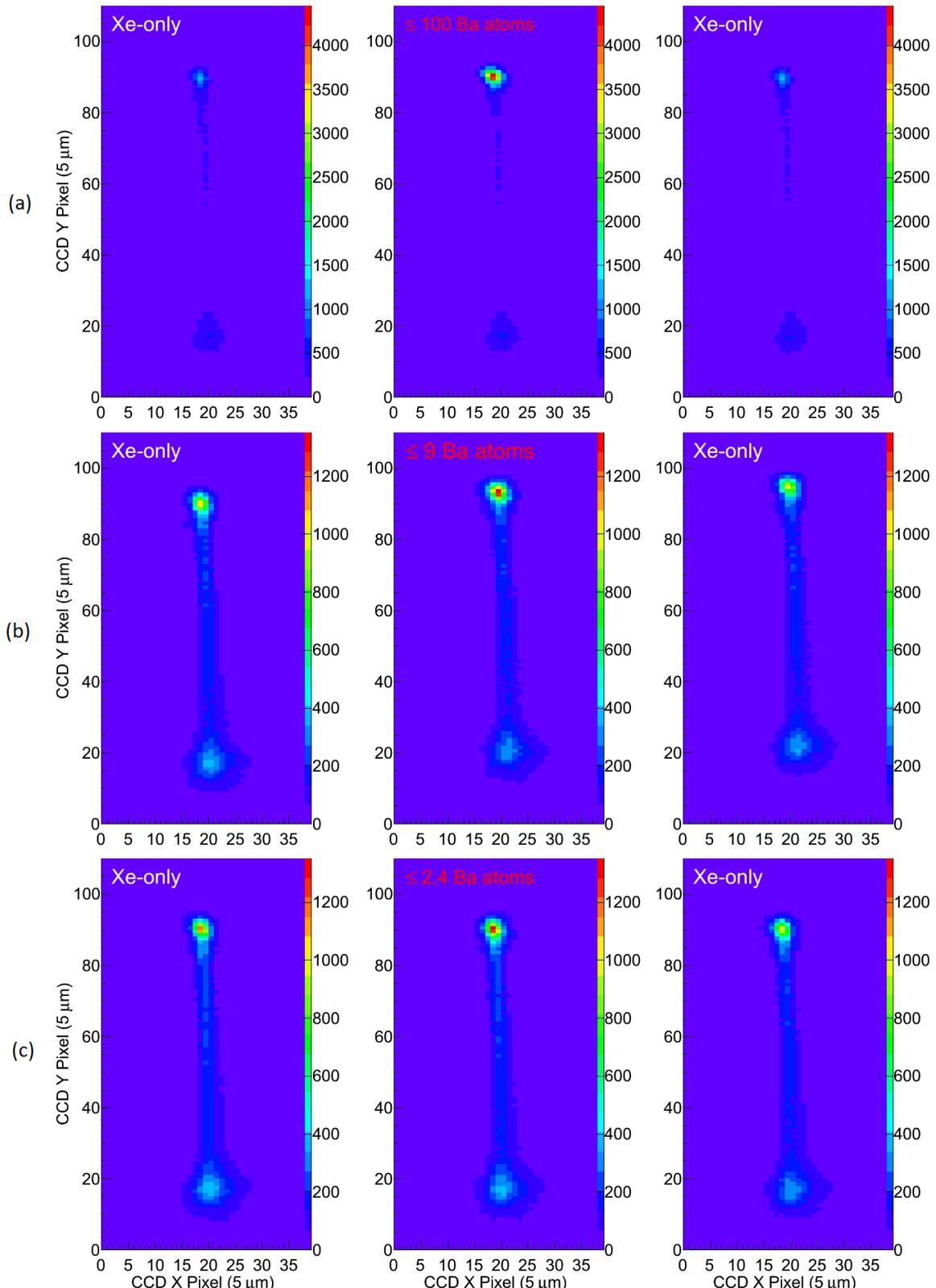


FIGURE 4.22. Raw images of three Ba^+ deposits yielding (a) ≤ 100 , (b) ≤ 9 , and (c) ≤ 2.4 Ba atoms, with their preceding and succeeding Xe-only deposits.

however positive signal after subtraction, even at the single-atom level, demonstrates that this variation is sufficiently low.

Xe-only images were also directly subtracted to produce images of 619-nm Ba fluorescence. In cases where the image has shifted slightly between runs, a binning redistribution was applied to do sub-pixel image shifting for better subtraction, though images which did not require this were preferred. 619-nm fluorescence images for several deposits of varying numbers of atoms are shown in Fig. 4.21.

It is important to note that no Ba signal is left behind after a sample is evaporated, even for large Ba^+ deposits. Raw images (pedestal-subtracted) of Ba^+ deposits and their preceding and succeeding Xe-only deposits are shown in Fig. 4.22 for several runs. The lack of historical buildup of Ba is important for the implementation of this method of Ba tagging on a probe in nEXO.

4.3.4. FURTHER CHECKS ON 619-NM PEAK. The 619-nm peak, as well as the 670-nm peak, was attributed to neutral Ba by a few further tests. A large Ba^+ deposit is compared to a deposit made with the Ba getter in Fig. 4.23(a). The 619-nm and 670-nm peaks were observed with both sources, and since the getter produces only neutral Ba, they were attributed to neutralized ions in Ba^+ deposits. Observation of the fluorescence with two different types of Ba sources is itself positive. In addition, observation of the fluorescence with the low deposit energy of the getter may bode well for the prospect of grabbing on a probe in LXe. Observation of a large deposit of Ar^+ in SXe is also shown in Fig. 4.23(a). The lack of fluorescence further disfavors a matrix-damage-related source of the fluorescence, such as color centers. Finally, to rule out the possibility that the signal in Ba imaging experiments is due to passed wavelengths far from the 620-nm band-pass region, e.g. infrared, imaging

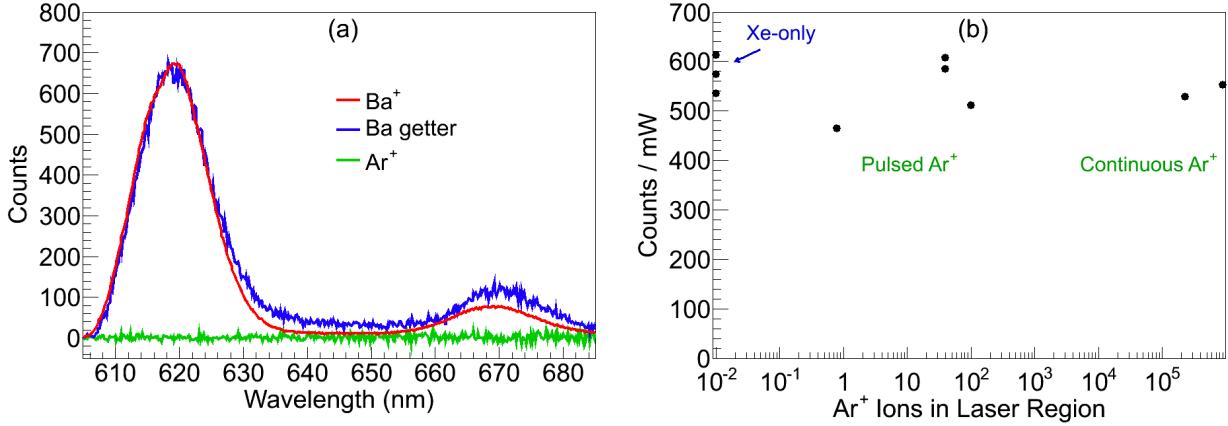


FIGURE 4.23. Comparison of signal observed for deposits with the Ba^+ ion beam (red), Ba getter (blue), and Ar^+ ion beam (green) (a), and signal through 620-nm band-pass from deposits of small to large numbers of Ar^+ ions in SXe.

experiments of Ar^+ deposits in SXe were performed with both pulsing and continuous ion beams. The same 620-nm band-pass filter was used, as well as the same focused 570-nm laser, to be similar to Ba^+ 619-nm imaging experiments. Summed counts from these deposits were consistent with background, as shown in Fig. 4.23(b).

4.4. SCANNED IMAGES

4.5. CANDIDATE Ba^+ LINES/BLUE EXCITATION

Since the fraction of neutralization of Ba^+ deposited in SXe is not known to be 100%, in this system and particularly in future tests of grabbing out of LXe, detection of single Ba^+ in SXe is still of interest. Preliminary studies of fluorescence from Ba^+ deposits were performed by blue laser excitation with the C480 dye laser.

Blue excitation of Ba^+ in SXe were first explored in the thesis of Shon Cook, wherein a set of sharp emission peaks were observed at 522, 575, 637, 712, and 814 nm, in decreasing amplitude. These peaks were attributed to emission from different vibrational states of a molecule composed of Ba and one or more H atoms [22]. This attribution is supported by

two experiments. Firstly, a reduction of those peaks is observed in deposits made at 44 K, well above the H₂ freezing temperature of 12 K, vs. deposits made at 11 K, as shown in Fig. [fig BaHx](a) *cite baspec in caption*, where the 522- and 575-nm peaks are observed. Secondly, the peaks are much stronger in deposits made with lower leak rates, as shown in Fig. [fig BaHx](b), for deposits made at 11 K. This is explained by a higher concentration of H₂ impurities in a matrix formed with a lower leak rate.

In contrast to lower BaH_x emission, several other emission peaks become prominent in deposits made at 50±5 K and observed at 11 K. A representative spectrum with fits is shown in Fig. [fig blues](a), and excitation spectra from those fits over the C480 dye range are shown in Fig. [fig blue](b). Of particular interest are the peaks at 530 nm and 630 nm. These peaks have identical excitation spectra, as well as identical bleaching rates, shown in Fig. [fig 530/630 bleach], indicating that they are due to the same excitation transition. Thus the 530- and 630-nm peaks may be due to relaxation to the ground and metastable D states, respectively, from the excited P state of Ba⁺ in SXe. However, matrix effects would need to be responsible for (a) the much slower bleaching rate than would be observed by optical pumping of Ba⁺ in vacuum, and (b) the higher number of counts observed in the 630-nm peak vs. the 520-nm peak, whereas the P → S transition is about 4× more likely than the P → D transition in vacuum.

CHAPTER 5

CONCLUSIONS

5.1. FUTURE STEPS

Talk about whatever Chris will do next ... ions? Other sites? Populations?

Talk about Adam's work, show pics and stuff.

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