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Ultrasonic detection of radioactive cesium isotopes using a magneto-optical trap

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ABSTRACT

We report the first magneto-optical trapping of radioactive ^{135}Cs and ^{137}Cs and a promising means for detecting these isotopes to ultrasensitive levels by a system coupling the magneto-optical trap (MOT) to a mass separator. The mass separator efficiently delivers a 20 kV ion beam of either isotope into a quartz-cube MOT cell having in one corner a small-diameter Zr foil, on which the ion beam is focused and into which the ions are implanted. Inductive heating of the foil releases ~45% of the implanted atoms into a MOT that uses large diameter beams and a dry-film coating to capture 3% of the released vapor. MOT fluorescence signals were found to increase linearly with the number of foil-implanted atoms over a range of 10^4 to 10^7 in trapped-atom number. The measured slope of MOT signal versus number implanted was equal in the case each isotope to within 4%, signifying our ability to measure $^{137}\text{Cs}/^{135}\text{Cs}$ ratios to within 4%. The isotopic selectivities of the mass separator and MOT combine to suppress interfering signal from ^{133}Cs by a factor of greater than 5×10^{12} in the case of detecting ^{135}Cs or ^{137}Cs . Our present sample detection sensitivity is one million atoms.

Keywords: magneto-optical trap, mass separator, cesium, ultrasensitive detection, radionuclides

1. INTRODUCTION

The detection of trace levels of long-lived radionuclides has a broad range of interests. In particular are uses of the radioactive isotopes ^{137}Cs ($t_{1/2} = 30$ y) and ^{135}Cs ($t_{1/2} = 2.3 \times 10^6$ y) that have been introduced to the environment by worldwide fallout from nuclear tests and operations of nuclear-fuel processing plants. Cesium-137 has long been used as a tracer of soil loss and erosion of watersheds^{1, 2}, and the measured ratio of $^{137}\text{Cs}/^{135}\text{Cs}$ in sediment would provide a chronometer superior to that supplied by measurements of ^{137}Cs alone.³ Also, as daughter products of the chemically inert and comparatively short-lived ^{137}Xe ($t_{1/2} = 3.8$ m) and ^{135}Xe ($t_{1/2} = 9.1$ h), the purely anthropogenic ^{137}Cs and ^{135}Cs (at ratios of roughly 1:1 from thermal fission of ^{235}U [ref. 4]) are likely effluents and thus signatures of nuclear-fuel reprocessing, with the precise $^{137}\text{Cs}/^{135}\text{Cs}$ ratio indicative of neutron fluxes within the fissioning system.⁵ The ability to measure ratios of $^{137}\text{Cs}/^{135}\text{Cs}$ would then provide a means for verifying non-proliferation compliance.

From manageable sizes of environmental samples, ^{137}Cs can be detected to ultratrace levels (*e.g.* to levels of 10^8 atoms from ~50 l of sea water^{6, 7}) by counting events of its strong gamma-ray emission at 662 keV. Detecting the comparatively very long-lived ^{135}Cs through radioactive decay-counting methods, however, would require impractically large samples. The lack of gamma emission and sole release of low-energy beta particles (0.21 MeV β^-) from ^{135}Cs present further difficulties to low-level counting methods. Conventional mass spectrometers also typically cannot distinguish ^{135}Cs in its low abundance of 10^{-10} – 10^{-12} [refs. 3, 8] against the signal "tail" from natural levels of ^{133}Cs .

Isotopic selectivities of 10^{10} and greater, as generally needed in the detection of $^{135,137}\text{Cs}$, are usually the domain of accelerator mass spectrometry⁹ and laser-based methods such as resonant-laser ionization mass spectrometry^{8, 10} and, more recently, atomic traps.¹¹⁻¹³ All three methods have been applied to the detection of rare radionuclides.⁸⁻¹³ In our approach to cesium detection, a conventional mass separator delivers an isotopically separated beam to a high-efficiency magneto-optical trap¹⁴ (MOT) for detection and further isotopic discrimination. This coupled system of mass separator and MOT presently achieves a system efficiency of 10^{-3} , a precision of potentially 4% to the ratio $^{137}\text{Cs}/^{135}\text{Cs}$, a detection limit of 4×10^6

Cs atoms, and a selectivity of $>10^{12}$ to $^{135,137}\text{Cs}$ for a performance similar to the best available AMS and RIMS-based methods in ultrasensitive detection. The high isotopic selectivity is owed in part to the near-resonant spectroscopic processes governing the capture of atoms within a MOT, and the reader is referred to the many excellent reviews describing their operation.¹⁵⁻¹⁷ Our high-efficiency MOT, developed in synergy with needs for trapping short-lived isotopes of Rb,¹⁸⁻²¹ draws from the work of Stephens and Wieman.²²

2. MASS SEPARATOR AND MOT

Figure 1 shows an overview of the coupled system of mass separator and MOT. Cesium, with mass numbers 133, 135, and 137 in roughly the proportion 1000:1:1, was loaded as cesium carbonate (Cs_2CO_3) into a tantalum crucible for the mass-separator source. Dual electron-beam bombardment of the crucible, with one beam controlling the vaporization rate and the other the ionization rate, liberates Cs vapor from the carbonate source and produces Cs^+ . The Cs ions are extracted electrostatically, accelerated to 20 keV, and focused with a quadrupole triplet through a 90° bending magnet to the focal plane of the mass separator where a single mass is selected. A second quadrupole triplet focuses the mass-selected ions into the MOT cell, a quartz cube of 7.6 cm per side that is open to the mass separator through a 5-mm diameter hole centered along a vertical edge. The ion beam traverses the horizontal diagonal of the cell and is focused and implanted into a metal catcher foil located opposite the cell opening. After a suitable implantation period, the foil is inductively heated to release the collected atoms as a neutral (uncharged) vapor.¹⁸ Picoammeters are used to measure the ion current at both the catcher foil and at a 5-mm I.D. metal collimator that is positioned along the ion-beam axis just before the cell. The collimator and catcher foil are located coaxially along the ion-beam path, with the collimator serving to trim the diameter of the ion beam before it enters the cell. We steer and center the ion beam on the foil by maximizing the ion current measured at the foil while minimizing the ion current at the entrance collimator. Earlier studies showed this method of ion-beam steering to be easily capable of centering the focused ion beam of measured 1 mm diameter fully within the foil diameter of 6 mm.. Our previous studies determined that Rb atoms were

Fig. 1: Coupled mass-separator/MOT system, with detail of MOT at inset. The mass-separated ion beam is focused onto a catcher foil located within the cubic MOT cell. Atoms implanted in the foil are later released by inductive heating for capture by a high-efficiency MOT.

Fig 2: D₂-line structure of Cs.

Along the vertical axis, the cell is centered within a pair of annular electromagnetic coils (13.5 cm I.D.) in (nearly) an anti-Helmholtz configuration that provide a quadrupole-field gradient of 7 G/cm.

The distinction of a “high efficiency” MOT comes from our use of (1) large-diameter laser-trapping beams and (2) an inert dryfilm (anhydrous) coating over the cell interior. The former augments the loading rate of atoms into the trap, as the loading rate scales with the fourth power of the beam diameter.¹⁶ Through the 7.6-cm square windows of the cubic MOT cell, we pass 4.5 cm diameter beams ($1/e$ width), which are larger than the beam diameters of 1-2 cm typically used in MOTs. The dryfilm coating greatly minimizes the loss of Cs to the cell walls, ideally ejecting a Cs atom back to the vapor on each wall collision for another chance to be trapped. Wall collisions also serve to reduce the atom kinetic temperature to “room temperature” after release from the hot ($\sim 800^\circ\text{C}$) foil, thereby increasing the fraction of atoms having speeds within the range of capture by the MOT. Presently, we use the dryfilm coating SC-77 (Silar Laboratories). Approximately 3% of the atoms released from the foil are captured by the MOT, as we will later show.

2.1 MOT Operation

Stable cesium (^{133}Cs) was first captured in a MOT in 1989 [ref. 35] and has since become a staple of optical-trap research. However, to our knowledge, no other isotope of Cs (none of which are stable) has been trapped in a MOT prior to this work.¹³ Figure 2 shows an energy diagram relevant and in common to the optical trapping of isotopes $^{133,135,137}\text{Cs}$ within the hyperfine structure of the $6^2P_{3/2} \leftarrow 6^2S_{1/2}$ D₂ line, given that all three isotopes have a nuclear spin of $7/2$. Listed in Table 1 for each isotope is the centroid frequency (ν_c) and the energy (δ_F) of the hyperfine level (F) relative to the fictitious unsplit level, designated by a dashed line in Fig. 2. The energy $E(F' \leftarrow F'')$ of an absorption transition is then calculated from

$$E(F' \leftarrow F'') = \nu_c - \delta_{F''} + \delta_{F'}, \quad (1)$$

where superscripts of double- and single-prime assign the associated F to the lower and upper state, respectively. The ordering of hyperfine levels in transition labels used here is consistently F' before F'' , with the direction of the intervening arrow indicating whether the transition is in absorption (left arrow) or emission (right arrow). From Table 1 (constructed from refs. [25–30]) and Eq. (1), we determine the transition frequencies ν_{trap} and ν_{repump} that together effect a closed cycling transition as required for cooling and trapping. The trapping frequency, ν_{trap} , is tuned to the main cycling transition of $5 \leftarrow 4$ but

ionized and transported to the foil with an efficiency of 35% [ref. 18], and we expect this efficiency applies as well to the case of Cs atoms.

The catcher foil consists of Zr of $6\text{ mm } \phi \times 25\text{ }\mu\text{m}$ thick backed by a tungsten disc of equal diameter and $50\text{ }\mu\text{m}$ thick. A short molybdenum wire ($180\text{ }\mu\text{m } \phi$) suspends the catcher foil within the cell and conducts the ion current outside the cell for measurement. As Fig. 1 indicates, the foil resides within a cylindrical quartz port of 20 mm O.D., 7 mm I.D. and 2.5 cm long, around which a coil is wound and powered at $\sim 7\text{ MHz}$ (and $\sim 20\text{ W}$) for $\sim 10\text{ s}$ to inductively heat the foil and release the implanted atoms. Roughly 45% of the implanted atoms are released when this Zr-overlaid catcher foil is heated to 800°C . We assume, from earlier studies on the release of Rb atoms,²³ that the majority of the released Cs atoms are neutral (un-charged) atoms.

Our high-efficiency MOT, described previously,¹⁸ uses three retroreflected pairs of mutually orthogonal laser beams directed horizontally and vertically along the face-normal axes of the quartz cell.

Table 1: Constants for calculating $^{133,135,137}\text{Cs } 6^2P_{3/2} \leftarrow 6^2S_{1/2}$ transition energies, for use with Eq. (1) and Fig. 2. References for values are in square brackets.

		133	135	137
	ν_c	351 725 718.50 MHz [25]	351 725 682.1 MHz [25, 28]	351 725 571.1 MHz [25, 28]
$6^2P_{3/2}$	δ_5	263.81 MHz [26]	280.93 MHz [30]	292.20 MHz [30]
	δ_4	12.82 MHz [26]	9.88 MHz [30]	10.35 MHz [30]
	δ_3	-188.44 MHz [26]	-200.66 MHz [30]	-208.71 MHz [30]
	δ_2	-339.64 MHz [26]	-354.90 MHz [30]	-369.27 MHz [30]
$6^2S_{1/2}$	δ_4	4021.78 MHz [27]	4254.26 MHz [29]	4425.54 MHz [29]
	δ_3	-5170.86 MHz [27]	-5469.76 MHz [29]	-5689.99 MHz [29]

occasionally excites $4 \leftarrow 4$, which subsequently removes atoms from the main cycling transition upon spontaneous emission of $4 \rightarrow 3$. A second (or repump) frequency, ν_{repump} , tuned to $4 \leftarrow 3$ then returns atoms back to the main cycling transition following their spontaneous emission of $4 \rightarrow 5$.

2.2 Laser Setup

As shown in Fig. 3 two cw Ti:sapphire lasers (Coherent 899-21), pumped by a single large-frame Ar⁺ laser (Coherent Innova), separately provide the trapping and repump light. The “trapping” and “repump” lasers have output powers of 1.2 and 0.3 W, respectively. A small portion of each laser output is passed to a wavemeter (Burleigh WA-100) for setting the nominal wavelength and to a spectrum analyzer (Coherent 240) for monitoring mode quality. Another portion of each laser output is sent to a reference-frequency circuit consisting of a double-pass acousto-optic modulator (AOM), a single-pass AOM, an electro-optic modulator (EOM), and a ^{133}Cs vapor cell that is 7.5-cm long and maintained at 10 C. Separate beams, one passing through the EOM and onto a photodiode detector and the other through the single-pass AOM, are sent counterpropagating across the cell to obtain saturated, Doppler-free absorption spectra in the manner of Hall *et al.*³¹ Figures 4a and 4b show the saturated-absorption spectra of ^{133}Cs obtained for transitions within $F' \leftarrow 4$ and $F' \leftarrow 3$, respectively. The features, appearing as first derivatives of resonance lines, are labeled by transition. Cross-over transition³² to frequencies between two upper levels (F'_1 and F'_2) are designated by $F'_1, F'_2 \leftarrow F''$.

Fig. 3: Arrangement of lasers and frequency-lock circuits producing the trapping and repump light for the MOT. Legend: AOM, acousto-optic modulator; EOM, electro-optic modulator; SP, single-pass use of the AOM; DP double-pass use of the AOM; PBS, polarizing cube beamsplitter.

We actively stabilize the laser frequency against specific zero-crossing points, or lock points, in the saturation spectra. Zero crossings in the $F' \leftarrow 4$ spectrum (Fig. 4a) are used to lock the trapping laser, and crossings in the $F' \leftarrow 3$ (Fig. 4b) spectrum are used to lock the repump laser. Vertical lines in Fig. 4 mark the trapping ($5 \leftarrow 4$) and repump ($4 \leftarrow 3$) frequencies needed for ^{135}Cs and ^{137}Cs MOTs. Owing to isotope shifts, the trapping and repump frequencies are different for each isotope. We also note that the trapping and repump frequencies needed for ^{135}Cs and ^{137}Cs are not coincident with the available lock-point frequencies of the ^{133}Cs spectra. The difference (Δ) between the needed laser frequency and a nearby lock-point frequency is precisely covered by the two AOMs in each reference-frequency circuit. The lock points used in the trapping of each isotope are labeled in Fig. 4 and were chosen to have Δ nominally equal for ^{135}Cs and ^{137}Cs , with the practical advantage of being able to use the same set of AOMs, only tuned slightly differently, for each isotope. We switched between trapping ^{135}Cs and ^{137}Cs by adjusting only the frequencies of the double-pass AOMs, from which the beams emerged un-deviated by changes in AOM frequency. Cesium-133 was trapped by using the retroreflected carrier passing through the double-pass AOM arrangement. The AOM settings provided in the case of trapping all isotopes a trapping frequency nominally shifted from resonance of $5 \leftarrow 4$ by -18 MHz , which we found to be the optimal detuning for operation of our Cs MOT. Specific detunings in the case of trapping ^{135}Cs and ^{137}Cs were measured as $-17.6 \pm 0.3\text{ MHz}$ and $-18.2 \pm 0.3\text{ MHz}$, respectively, with reference to line center at conditions in the MOT.

3. MOT-SIGNAL MEASUREMENTS AND ANALYSES

Fluorescence from the cloud of atoms trapped in the MOT was collected off-axis through a cell window at $f/7.5$ (for a collection solid angle of $\Omega = 0.014\text{ sr}$) and focused onto an avalanche photodiode (APD) of responsivity $R = 1.72\text{ mV/nW}$ at 852 nm , as measured after a transimpedance amplifier. A narrow-band interference filter (10 nm fwhm about 850 nm , 50% transmission) and a $1\text{-mm } \phi$ field stop at the APD helped minimize unwanted signals from background and scattered light. Still, a small constant signal persisted from laser light scattered off the cell walls. We dithered the frequency of the trapping laser, thus

Fig. 4: Saturated absorption spectra of the $6^2P_{3/2} \leftarrow 6^2S_{1/2}$ transitions of ^{133}Cs for (a) $F' \leftarrow F'' = 4$ and (b) $F' \leftarrow F'' = 3$. Trapping ($5 \leftarrow 4$) and repump ($4 \leftarrow 3$) frequencies for ^{135}Cs and ^{137}Cs are labeled and located by bold vertical lines. Circled zero-crossings of the spectra are used for laser-frequency stabilization.

modulating the cloud fluorescence, in order to discriminate by coherent detection the trapped-cloud fluorescence from this constant background. The laser frequency was dithered ± 10 MHz about the trapping frequency by at a rate of 170 Hz, producing an approximately 65% modulation of the fluorescence signal, and the modulated output of the APD was analyzed and demodulated by a lockin amplifier. The demodulated APD signal was converted to the peak of the modulated fluorescence signal (S_F) through measured calibration factors. Under the reasonable assumption of a two-level atom, S_F is related to the number of trapped atoms (N_T) by¹⁶

$$S_F = N_T (\Omega/4\pi) R C \left(\frac{\Gamma}{2} \right) \frac{(I/I_{\text{sat}})}{1 + 4(\delta/\Gamma)^2 + (I/I_{\text{sat}})}, \quad (2)$$

where $\Gamma = 3.288 \times 10^7 \text{ s}^{-1}$ [ref. 33] is the spontaneous emission rate of Cs $6P_{3/2}$ (assumed equal for all isotopes), $\Omega/4\pi$ is fractional collection of fluorescence over 4π sr, δ is the detuning of the trapping-laser frequency from resonance of the $5 \leftarrow 4$ trapping transition, I_{sat} is the saturation irradiance of the trapping transition, and C accounts for measured transmission losses at 852 nm through the cell, the lens, and the narrow-band interference filter placed before the APD. The aggregate three-dimensional light field of the MOT is assumed to illuminate the trapped atoms isotropically by all polarizations, for which we calculate $I_{\text{sat}} = 2.71 \text{ mW/cm}^2$ for the $6P_{3/2}(F'=5) \leftarrow 6S_{1/2}(F''=4)$ transition of Cs.

During the course of measurements, we estimate that laser power and frequency were constant with random variations of $\pm 5\%$ and ± 0.3 MHz, respectively. By Eq. (2), these fluctuations in I and δ combined to yield a source of random error in N_T of $\pm 10\%$ ($\pm 2\sigma$) as determined from the measured fluorescence signal S_F . The proportionality between S_F and N_T in Eq. (2) presumes the trapped cloud of atoms is optically thin over frequencies of the resonance-fluorescence transition $5 \rightarrow 4$, an assumption we have supported through calculations of radiative transport. For our conditions, we estimate that corrections to N_T for radiative escape remain well within our random measurement uncertainty for N_T of $\pm 10\%$ up to the largest number of atoms trapped of 8×10^6 .

3.1 MOT Signal Following Foil Release

The solid line of Fig 5a shows the MOT fluorescence signal of ^{135}Cs obtained as ^{135}Cs is released from the catcher foil during a ~ 9 s heating pulse (Fig. 5b). In Fig. 5a, the raw MOT fluorescence signal from the lockin amplifier (operating with an integration time of 30 ms) has been converted to the number of trapped atoms by Eq. (2). The rise and decay of MOT fluorescence signal, peaking for the data of Fig. 5a at an equivalent number of trapped atoms of 5.4×10^6 , occur as the released atoms are captured in the MOT and then eventually expelled, primarily by collisions with the background gas. Figure 5b shows the foil temperature during the heating pulse as measured by an optical pyrometer with a 10 Hz frequency response. The minimum temperature measurable by the pyrometer of 400°C is indicated in Fig. 5b by the dashed horizontal line. Both the MOT and pyrometer signals were sampled by analog-to-digital converters at 10 Hz, which was sufficiently fast to capture the profiles of MOT fluorescence and foil temperature of time constants > 2 s. Shown by the dashed line in Fig. 5a is the fluorescence signal we observed during the heated release of ^{133}Cs from the foil. No ^{133}Cs atoms were MOT-captured as the lasers were still tuned to trap ^{135}Cs , but the coincidence of the (modulated) trapping-laser frequency with the Doppler-broadened $5,4,3 \leftarrow 4$ feature of ^{133}Cs generated a weak burst of fluorescence signal as the foil-released vapor quickly filled and exited the cell. The raw ^{133}Cs fluorescence signal was scaled as an equivalent number of trapped atoms, in order to interpret its magnitude as an interference to the number of trapped atoms determined from the ^{135}Cs MOT signal. As will be explained, we use the ^{133}Cs fluorescence signal to (1) represent the temporal envelope of atoms released into and evacuated from the cell and (2) determine the isotopic detection selectivity of ^{135}Cs over ^{133}Cs . Zero time for both traces of Fig. 5a was set at the instant of foil heating. The foil-temperature pulse associated with the dashed line of Fig. 5a is not shown but is essentially identical to that of Fig. 5b.

Fig. 5: (a) MOT-signal envelope when trapping ^{135}Cs (solid line) compared with the weak fluorescence burst (dashed line) from the release of 2.5×10^{13} ^{133}Cs atoms. In both cases, the MOT was tuned to trap ^{135}Cs , and $t = 0$ was set relative to the first inflection of the (b) foil temperature. A crosshair intersects the fluorescence-burst at the time of peak MOT signal to indicate its interference magnitude. The pyrometer monitoring the foil temperature responds for 400 °C and greater.

During the period of ion implantation prior to atom release, the ion current (i) at the catcher foil was steady and was measured with a picoammeter for later integration to yield the number of implanted atoms (N_i). To convert integrated current $\int i dt$ to N_i , we used a measured factor of 2.5 for secondary electron release³⁴ to account for an additional current owed to the release of electrons from the Zr-W catcher foil on bombardment by the 20 kV ion beam. The number of implanted atoms were then calculated from $N_i = (\int i dt) / (1.6 \times 10^{-19} / (1 + 2.5))$. For example the solid line of Fig. 5a followed from the implantation of $^{135}\text{Cs}^+$ into the foil and is associated with a measured integrated current drawn from the catcher foil of 220 pC, which we convert (as noted in the figure) to 3.9×10^8 implanted atoms of ^{135}Cs . The dashed line followed from the implantation of 1.8×10^{13} ^{133}Cs atoms.

3.2 Vapor Fluorescence Signal and Cell-Coating Performance

Before the foil temperature (Fig. 5b) reaches a plateau of ~ 800 °C, the vapor fluorescence signal (dashed line, Fig. 5a) is seen to peak quickly and then begin a roughly exponential decay with a time constant of approximately 1 s, as limited by the release of atoms from the foil. The rising edge of the vapor signal is controlled by the evacuation rate of atoms from the cell. An evacuation time constant of $\tau_{1/e} \approx 0.3$ s was determined from a simulation of the vapor fluorescence signal, using 1 s for the foil-release time constant. The release of atoms from the foil and their evacuation from the cell is essentially complete after 5 s of foil heating, but the foil remains heated for an additional ~ 4 s as a continued purge of a residual amount of implanted atoms. The heated foil also continually repels the released vapor and therefore does not contribute to the adsorption loss of atoms. The estimated evacuation time of 0.3 s is consistent with atom loss through the two 5-mm ϕ open ports of the cell (and through a minor annular gap to uncoated surfaces behind the foil), indicating that few atoms are lost by adsorption to the SC-77 coated cell surface.

3.3 Isotopic Selectivity

The crosshair in Fig. 5a marks the time of peak ^{135}Cs MOT signal and the corresponding amount of ^{133}Cs vapor that would be present if released simultaneously with ^{135}Cs from the foil. As shown, the peak ^{135}Cs MOT signal of 5.4×10^6 atoms coincides with an interfering signal of $\sim 3 \times 10^3$ atoms from a remaining small

Fig. 6: Raw data for the repeated cycles of (1) implanting ions from the mass separator into the catcher foil, (2) MOT-capture of ^{135}Cs atoms following their heated-foil release, and (3) heating the foil and MOT-capturing atoms again to measure a background release of atoms. Top panel: ^{135}Cs MOT signal. Bottom panel: foil-heating pulses. MOT signal converted to the number of trapped atoms by Eq. (2).

amount of ^{133}Cs vapor. Given that this potential interference was the result of implanting 1.8×10^{13} atoms of ^{133}Cs , compared with an implanted 3.9×10^8 atoms of ^{135}Cs , we estimate our MOT-based detection method to have a selectivity of $>5 \times 10^7$ for the detection of ^{135}Cs in the presence of ^{133}Cs . When compounded by a measured isotopic selectivity of 10^5 of the mass separator,³⁴ the overall suppression of ^{133}Cs is expected to exceed 5×10^{12} . For MOT signals acquired after the time of peak signal, we note that interference from ^{133}Cs is further suppressed, though at some expense of signal and thus a reduced signal-to-noise ratio. The isotopic selectivity for the MOT detection of ^{137}Cs was similarly tested and found to exceed 1×10^9 in the presence of ^{133}Cs . The interference of ^{133}Cs to ^{137}Cs signals is suppressed an additional five orders of magnitude by the selectivity of the mass separator, yielding an ultimate selectivity of $\geq 1 \times 10^{14}$.

4. DETECTION LINEARITY AND SYSTEM EFFECIENCY

Figure 6 shows data obtained in the case of trapping ^{135}Cs during an extended sequence of (1) implanting ^{135}Cs ions from the mass separator into the MOT catcher foil, (2) heating the foil to release the atoms and capturing the released plume in the MOT, and (3) heating the foil and MOT-capturing atoms for a second time to determine the signal level of background release. The individual events of ion implantation and foil heating are labeled in Fig. 6. The bottom panel shows the foil temperature (measured with an optical pyrometer), and the top panel shows the raw fluorescence signal converted to the number of trapped atoms by Eq. (2). For the data of Fig. 6, the measured 6-beam irradiance was 20 mW/cm^2 for $I/I_{\text{sat}} = 7.4$. During implantation periods, the ion currents were steady and were measured with a picoammeter for later integration. As mentioned, the integrated current was corrected for secondary electron emission to yield the number of implanted atoms. The ion current was either raised or lowered between successive releases by adjusting the source strength of the mass separator. Ultimately, the number of implanted atoms (N_i) was varied over a factor of about 500, producing a similar variation in the peak number of atoms trapped

Fig. 7: Measurements of the peak number of atoms trapped following foil release versus the number of atoms implanted in foil. All measurements for ^{135}Cs (open circles) and ^{137}Cs (filled circles) include vertical error bars of $\pm 2\sigma$ of typically $\pm 10\%$ of the measurement magnitude; error bars not visible are within the symbol height. The solid line represents the least-squares linear fit to all measurements.

(N_T^{peak}). Data for the case of trapping ^{137}Cs was obtained by the same sequence shown in Fig. 6 and was analyzed similarly. For trapping either isotope, the background release from the foil represented a roughly constant level of 2×10^4 trapped atoms and could be subtracted with an uncertainty ($\pm 2\sigma$) as small as $\pm 4 \times 10^3$ atoms.

Measurements of N_T^{peak} versus N_I are shown for both isotopes in Fig. 7, where the vertical axis represents the peak number of atoms trapped during the first foil release following implantation, as corrected for the level of background release. Measured values are indicated by symbols—open for ^{135}Cs and filled for ^{137}Cs —and vertical bars represent the $\pm 2\sigma$ measurement uncertainty, including errors in subtracting the background level and the random error in determining the number of implanted atoms as translated to N_T . The solid line represents the least-squares linear fit to all measurements in the log-linear form of $\ln(N_T^{\text{peak}})$ versus N_I , which gave a more equal weighting in the χ^2 minimization to all measurements obtained over the nearly three-decade range in N_T . The fitted slope of 0.0142 ± 0.0006 (error of $\pm 2\sigma$) signifies that of the atoms implanted in the foil, 1.4% are released and captured in the MOT. The true slope is affected by the value of the secondary electron coefficient used to calculate the number of implanted atoms (the abscissa of Fig. 7) from the integrated current. Given the measured secondary electron emission coefficient of 2.5 ± 0.5 [ref. 34], the efficiency for the release and MOT-capture of atoms is assigned as $1.4 \pm 0.2\%$. As a product of the foil-release and MOT-capture efficiencies, with the former being $\approx 45\%$ [ref. 23], the measured slope of Fig. 7 indicates the MOT captures $\approx 3\%$ of the released atoms. When multiplied with the previously mentioned efficiency for atom transfer from source to foil of 35% (inclusive of ionization, mass separation, and foil implantation), the slope from Fig. 7 yields an overall transfer efficiency of 0.5% from sample to MOT.

5. DISCUSSION OF ISOTOPE-RATIO APPLICATION

An accurate measurement of the isotope ratio by this method can be reduced without prior calibration to the simple ratio of peak MOT signals (S_F^{peak}) if (1) S_F^{peak} is strictly proportional to N_I and (2) the proportional constant between S_F^{peak} and N_I is isotope independent. From the excellent linearity displayed by the

measurements of Fig. 7, the first criterion appears to be met over a wide dynamic range from several million to, in principle, several trapped atoms. The second concerns possible isotopic differences in efficiency with which atoms get released from the foil, captured by the MOT, and detected. Ideally, the data of Fig. 7 for the separate cases of detecting ^{135}Cs and ^{137}Cs would yield identical slopes (m) when least-squares fit to a straight line. We find this ratio of best-fit slopes—determined as m_{137}/m_{135} —to be nearly unity at 1.04 ± 0.11 (error of $\pm 2\sigma$). The 4% departure from unity we take as representative of systematic errors that could arise in the assumption of isotopic independence, although the uncertainty in the slope ratio does admit the ideal value of unity. The random error in the slope ratio of 11% reflects our present degree with which we account for factors that influence MOT performance and could likely be reduced through continuous and more precise monitoring of drifts in laser power and laser beam-parameters. We note that the estimated error of 4% for an isotope-ratio measurement applies only to procedures in the MOT-based detection method. In practice, particularly at ultratrace levels of the isotopes when N_I will not be known or measurable, the uncertainty in a measured isotope ratio will also involve the operational stability of the mass separator.

6. SUMMARY

We applied our system of a coupled mass separator and MOT to the first magneto-optical trapping of ^{135}Cs and ^{137}Cs and examined the system's performance in the detection of these ultratrace radioactive isotopes. Atoms (as ions) from the mass separator were focused, implanted, and concentrated on a small catcher foil within the MOT cell. Upon foil heating, the implanted atoms were released for capture by the MOT. Approximately 1% of the implanted atoms were captured by the MOT, where they were detected quantitatively in fluorescence. We recorded the temporal envelope of MOT signals following the release of between 1×10^6 to 6×10^8 implanted atoms (N_I) and found the peak MOT signal (S_F^{peak}) to vary linearly with N_I over the corresponding range in the peak number of atoms trapped (N_T^{peak}) of 2×10^4 to 8×10^6 . In principle, the linearity between N_T^{peak} and N_I extends as well to the ultrasensitive level of several trapped atoms. The proportional chain of $S_F^{\text{peak}} \propto N_T^{\text{peak}} \propto N_I$ permits a direct measurement of the isotope ratio from the ratio of peak fluorescence signals. While isotope ratios were not expressly measured here, we found the slope of S_F^{peak} versus N_I was equal for the case of each isotope to within 4%, signifying the potential for accurate measurements of $^{137}\text{Cs}/^{135}\text{Cs}$ through simple ratios of S_F^{peak} . The MOT-based detection suppressed interfering signals from ^{133}Cs atoms by factors of $>5 \times 10^7$ and $>1 \times 10^9$ in cases of detecting ^{135}Cs and ^{137}Cs , respectively. Including the isotopic selectivity of $>10^5$ of our mass separator, the overall suppression of ^{133}Cs exceeds 5×10^{12} .

Our somewhat high MOT detection limit of 4×10^3 trapped atoms is limited primarily by a low-level background release of atoms that we are presently seeking to reduce. Nonetheless, when combined with an overall transfer efficiency from source to MOT of 0.5%, this MOT detection limit provides a sample sensitivity of 8×10^5 Cs atoms that compares favorably with sensitivities achieved by AMS- and RIMS-based systems. We have traced the background-signal source to the 25- μm Zr catcher foil itself, wherein we believe the unreleased atoms (a substantial ~50% of the implanted atoms) accumulate, diffuse, and later release. We are currently testing the performance of much thinner Zr foils, which should significantly reduce the volumetric retention of atoms in the foil and thereby improve the MOT detection limit.

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REFERENCES

1. W. Pennington, R. S. Cambray, and E. M. Fisher, *Nature* **242**, 324 (1973).
2. J. C. Ritchie and J. R. McHenry, *J. Environ. Qual.* **19**, 215 (1990).
3. T. Lee, T-L Ku, H-L Lu, and J-C Chen, *Geochim. Cosmochim. Acta* **57**, 3493 (1993).
4. E. A. C. Crouch, *Atomic Data and Nuclear Tables* **19**, 417 (1977).
5. J-H Chao and C-L Tseng, *Nucl. Instrum. Methods A* **372**, 275 (1996).
6. M. N. Alam, M. I. Chowdhury, Masud Kamal, S. Ghose, N. Mahmood, A. K. M. A-Matin, and S. Q. Saikat, *Appl. Radiat. Isot.* **47**, 33 (1996).
7. M. Aoyama, K. Hirose, T. Miyao, and Y. Igarashi, *Appl. Radiat. Isot.* **47**, 159 (2000).
8. K. Wendt, "Ultratrace Determination of Long-Lived Radioactive Isotopes," in *Resonance Ionization Spectroscopy, Ninth International Symposium*, AIP Conference Proceedings 454, American Institute of Physics, Woodbury, NY, pp. 39-46, 1998.
9. L. K. Fifield, *Rep. Prog. Phys.* **62**, 1223 (1999).
10. L. Pibida, J. M. R. Hutchinson, J. Wen, and L. Karam, *Rev. Sci Instrum.* **71**, 509 (2000).
11. C. Y. Chen, Y. M. Li, K. Bailey, T. P. O'Connor, L. Young, and Z.-T Lu, *Science* **286**, 1139 (1999).
12. K. Bailey, C. Y. Chen, X. Du, Y. M. Li, Z.-T Lu, T. P. O'Connor, and L. Young, *Hyperfine Interactions* **127**, 515 (2000).
13. D. J. Vieira, S. G. Crane, R. Guckert, and X. Zhao, *Hyperfine Interactions* **127**, 387 (2000).
14. E. L. Raab, M. Prentiss, A. Cable, S. Chu, and D. E. Pritchard, *Phys. Rev. Lett.* **59**, 2631 (1987).
15. H. Metcalf and P. van der Straten, *Phys. Rep.* **244**, 203 (1994).
16. C. S. Adams and E. Riis, *Prog. Quant. Electr.* **21**, 1 (1997).
17. V. I. Balykin, V. G. Minogin, and V. S. Letokhov, *Rep. Prog. Phys.* **63**, 1429 (2000).
18. R. Guckert, X. Zhao, S. G. Crane, A. Hime, W. A. Taylor, D. Tupa, D. J. Vieira, and H. Wollnik, *Phys. Rev. A* **58**, R1637 (1998).
19. X. Zhao, S. G. Crane, R. Guckert, and D. J. Vieira, *Phys. Rev. A* **60**, 4730 (1999).
20. S. G. Crane, X. Zhao, W. Taylor, and D. J. Vieira, *Phys. Rev. A* **62**, 011402(R) (2000).
21. S. G. Crane, S. J. Brice, A. Goldschmidt, R. Guckert, A. Hime, J. J. Kitten, D. J. Vieira, and X. Zhao, *Phys. Rev. Lett.* **86**, 2967 (2001).
22. M. Stephens and C. Wieman, *Phys. Rev. Lett.* **72**, 3787 (1994).
23. S. G. Crane, "Parity Violation Observed in the Beta-Decay of Magnetically Trapped ^{82}Rb Atoms," PhD Thesis, Utah State University, Report LA-UR-015199 (2001).
24. D. Sesko, T. Walker, C. Monroe, A. Gallagher, and C. Wieman, *Phys. Rev. Lett.* **63**, 961 (1989).
25. Th. Udem, J. Reichert, and T. W. Hansch, *Phys. Rev. A* **62**, 031801(R) (2000).
26. C. E. Tanner and C. Wieman, *Phys. Rev. A* **38**, 1616 (1988).
27. E. Arimondo, M. Ignusci, and P. Violino, *Rev. Mod. Phys.* **49**, 31 (1977).
28. C. Thibault, F. Touchard, S. Buttgenbach, R. Klapisch, M. De Saint Simon, H. T. Duong, P. Jacquinet, P. Juncar, S. Liberman, P. Pillet, J. Pinard, J. L. Vialle, A. Pesnelle, and G. Huber, *Nucl. Phys.* **A367**, 1 (1981).
29. G. H. Fuller, *J. Phys. Chem. Ref. Data* **5**, 835 (1976).
30. F. Ackermann, E. W. Otten, G. zu Pulitz, A. Schenck, and S. Ullrich, *Nucl. Phys.* **A248**, 157 (1975).
31. J. L. Hall, L. Hollberg, T. Baer, and H. G. Robinson, *Appl. Phys. Lett.* **39**, 680 (1981).
32. W. Demtröder, *Laser Spectroscopy*, Second Enlarged Edition, Springer, New York (1996).
33. L. Young, W. T. Hill III, S. J. Sibener, S. D. Price, C. E. Tanner, C. E. Wieman, and S. R. Leone, *Phys. Rev. A* **50**, 2174 (1994).
34. D. J. Vieira, M. D. Di Rosa, X. Zhao, S. G. Crane, R. Guckert, J. J. Kitten, W. A. Taylor, "High-Efficiency Mass Separator for use with Magneto-Optical Traps," *in preparation*.