

**Efficient isotope separation by single-photon atomic sorting**M. Jerkins,<sup>1</sup> I. Chavez,<sup>1</sup> U. Even,<sup>2</sup> and M. G. Raizen<sup>1</sup><sup>1</sup>*Center for Nonlinear Dynamics and Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA*<sup>2</sup>*Sackler School of Chemistry, Tel-Aviv University, Tel-Aviv, Israel*

(Received 7 May 2010; published 15 September 2010)

We propose a general and scalable approach to isotope separation. The method is based on an irreversible change of the mass-to-magnetic moment ratio of a particular isotope in an atomic beam, followed by a magnetic multipole whose gradients deflect and guide the atoms. The underlying mechanism is a reduction of the entropy of the beam by the information of a single scattered photon for each atom that is separated. We numerically simulate isotope separation for a range of examples, which demonstrate this technique's general applicability to almost the entire periodic table. The practical importance of the proposed method is that large-scale isotope separation should be possible, using ordinary inexpensive magnets and the existing technologies of supersonic beams and lasers.

DOI: [10.1103/PhysRevA.82.033414](https://doi.org/10.1103/PhysRevA.82.033414)

PACS number(s): 32.80.Xx, 32.10.Bi

The efforts to separate isotopes date back to the 1930s and fall into several categories. Two standard methods of separation are gaseous diffusion and the ultracentrifuge [1–3]. These methods require many stages of enrichment and are very inefficient. Furthermore, these methods are suitable only for a few elements that can be kept in gas phase, which is a common limitation of isotope separation schemes [4]. Isotope separation is also accomplished with mass spectrometry [5,6]. This method has high isotopic selectivity due to the use of a quadrupole mass filter, but it is very inefficient due to the low probability of electron-bombardment ionization and is limited by space charge. In recent years the method of isotope separation by laser ionization has been developed [7]. This approach is highly selective but requires multiple high-powered lasers for efficient ionization. The production rate is also limited by resonant charge exchange.

With this background, it is clear that there is an urgent need for a new and efficient method of isotope separation, the topic of our work. We first provide an overview of the approach and relate it to the historic problem of Maxwell's demon. We then analyze several representative cases using available NIST data [8] and provide the results of numerical simulations. Finally we discuss the prospects for scalability and experimental realizations.

We start with a generic prototype for isotope separation: a collimated atomic beam of a single element, composed of multiple isotopes. To be more specific, we consider a three-level atom with an initial ground state  $|i\rangle$ , an electronic excited state  $|e\rangle$ , and a final state  $|f\rangle$ .

We further assume that the magnetic moment of state  $|i\rangle$ ,  $m_i$ , is different than the magnetic moment of state  $|f\rangle$ ,  $m_f$ . Now suppose that an atom crosses a laser beam which induces an irreversible transition from state  $|i\rangle$  to state  $|f\rangle$  by absorption followed by spontaneous emission. The laser is tuned to one isotope, changing its magnetic moment, while not affecting the others. The atom then passes through a magnetic gradient  $\nabla B(x)$ , created by a magnetic multipole. The magnetic multipole acts like a filter, guiding only low-field seeking states.

We call this process single-photon atomic sorting because each atom is sorted by scattering one photon. The power of such a technique lies in its extremely efficient use of

laser power, which, when combined with a very bright atomic source, can produce a scalable, general, and relatively inexpensive method of isotope separation. It is closely related to a one-way barrier for atoms that was used as a general method for cooling the translational motion of atoms [9]. The goal is to lower the entropy of the atomic beam by separating the isotopes. This process can be viewed as a realization of Maxwell's demon in the sense proposed by Leo Szilard in 1929. Here the demon acts as a sorter, sending each isotope in a different direction. The entropy of the beam cannot be lowered with any time-dependent Hamiltonian such as an rf drive [10], and an irreversible step is required. The atom scatters one spontaneous photon from the laser beam, increasing the photon's entropy. This increase compensates for the decrease in the entropy of the beam.

We now discuss a more realistic scenario illustrated in Fig. 1. The starting point for this approach must be an atomic beam that has the lowest possible entropy of translational motion. Collimation of an effusive beam is not a viable approach since the resulting flux is too small [11]. The best candidate is the supersonic beam, which is generated with a high-pressure carrier gas expanding through a small aperture [12]. Supersonic beams possess remarkable properties, such as an angular divergence of only a few degrees and a velocity spread that is 1% of the mean velocity [13]. These beams are typically pulsed, but for the purpose of isotope separation they should be run quasicontinuously to maximize throughput.

Many elements are solid at room temperature and cannot be put directly into a supersonic nozzle. In order to apply our techniques generally to the entire periodic table, we propose a scheme that will enable atoms of any species to be entrained into a supersonic beam composed of an inert gas, such as a noble gas. We can achieve efficient entrainment using an annular oven, which is cylindrically symmetric around the supersonic nozzle with a small aperture perpendicular to the supersonic flow of atoms as illustrated in Fig. 1. A temperature gradient between the lower and upper sections of the oven produces a collimated effusive beam over the region intersected by the supersonic beam. The oven is lined with a stainless steel mesh, which wicks back atoms that are not entrained into the supersonic flow. The oven is, therefore, self-circulating in the sense that atoms not entrained will not

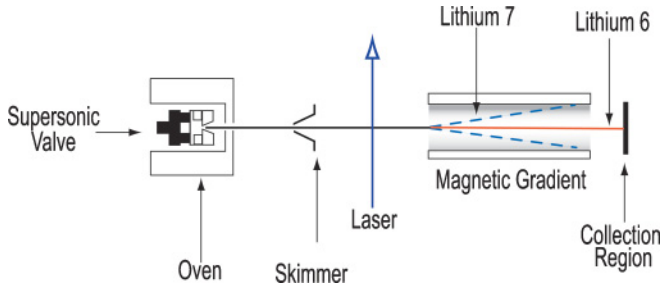


FIG. 1. (Color online) Schematic of the setup for isotope separation of lithium. Atoms from an annular oven are entrained into the flow of a carrier gas from a supersonic nozzle. A laser is tuned the  $D_2$  line of  ${}^7\text{Li}$  and populates all the atoms into the  $2^2S_{1/2} F = 1$  manifold, forcing them to be antiguided.

exit the oven and will have other chances to be entrained, as in continuous flow reflux ovens [14]. This self-circulation greatly reduces the initial amount of material needed for separation, as well as the background pressure and the need for recycling through vacuum pumps.

The entrainment we can achieve is as high as 5–10% of the initial beam flux because it occurs in an intermediate region in which the supersonic beam is no longer opaque, but there are still a large number of collisions, allowing the atoms to come into thermal equilibrium with the supersonic beam and continue cooling as the beam expands. Figure 2 shows simulation results of the entrainment of lithium into a supersonic beam of neon. We used the direct simulation Monte Carlo method to model the supersonic flow, which relies on the fact that the mean free path of the atoms is not negligible in order to solve the Boltzmann equation and compute the motion and collisions of representative atoms [15]. The simulation assumes an Even-Lavie valve, which has a beam flux of  $4 \times 10^{23}$  atoms/sr sec. That valve can be operated in a pulsed mode at 1 kHz, and, given available pumping speeds, the average continuous beam flux would be  $\sim 5 \times 10^{22}$  atoms/sr sec. Although this beam flux could probably be improved with

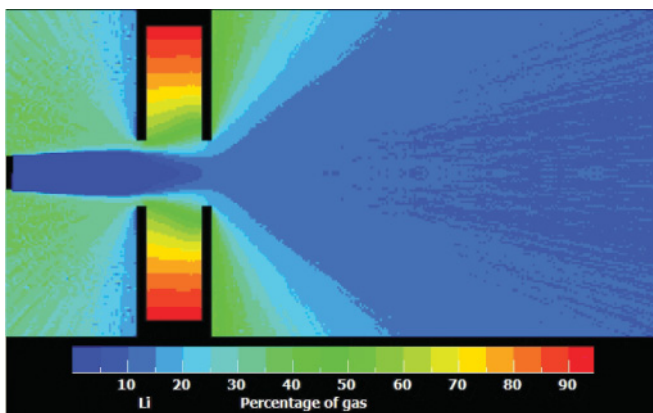


FIG. 2. (Color online) Direct simulation Monte Carlo results of the entrainment of lithium into a supersonic beam of neon using an annular lithium oven. This method is widely used to simulate rarified gas dynamics like those present in supersonic beams. The high 10% entrainment efficiency shown here enables isotope separation of significant quantities.

a nozzle designed specifically for maximum throughput, the following estimates will not assume any special modifications or optimizations. We intend to place a 5-mm diameter skimmer 15 cm from the nozzle, which implies that  $\sim 3 \times 10^{18}$  atoms/s will survive the skimmer. The 10% entrainment shown in Fig. 2 means that we expect  $\sim 3 \times 10^{17}$  lithium atoms/s. This beam flux is orders of magnitude higher than any atomic beam produced directly from an effusive oven [11]. The intense brightness afforded by utilizing a supersonic beam will allow this method of isotope separation to be applied in a large-scale industrial setting, which was previously unrealistic using traditional sources collimated from an oven.

After passing through the skimmer, the beam propagates into the laser region. The desired isotope will then undergo an irreversible change in magnetic moment, differentiating it from the other isotopes that are unaffected by the laser beam. After interacting with the laser, atoms proceed to enter a tube surrounded by multipole magnets, which produce a magnetic gradient that guides low-field seeking atoms and antiguides high-field seeking atoms [16]. Atoms which are forced toward regions of low (high) magnetic flux density are called low-field seeking (high-field seeking) atoms. The force due to the inhomogeneous magnetic field is

$$F = \mu_B g_J m_J \nabla B,$$

where  $\mu_B$  is the Bohr magneton,  $g_J$  is the Lande  $g$  factor,  $m_J$  is the projection of the total angular momentum on the quantization axis, and  $\nabla B$  is the gradient of the magnetic field. The maximum magnetic field of the gradient would be strong enough to cause a few elements, such as lithium, to enter the Paschen-Back regime. Most of the periodic table, however, would remain in the weak field limit because of the strong LS coupling present in heavier atoms.

Lithium serves as a simple example because it has two stable isotopes,  ${}^6\text{Li}$  and  ${}^7\text{Li}$ , with natural abundances of 7.6 and 92.4% respectively. In our proposed experiment, illustrated in Fig. 1, lithium is entrained into the supersonic beam, and a 670.96-nm laser tuned the  ${}^7\text{Li}$   $D_2$  line [ $2^2S_{1/2}(F = 2) \rightarrow 2^2P_{3/2}(F = 1 \text{ or } F = 2)$ ] optically pumps the  ${}^7\text{Li}$  into a high-field seeking state. The laser depletes the  $2^2S_{1/2} F = 2$  manifold and optically pumps all of the atoms into the  $2^2S_{1/2} F = 1$  manifold. At fields greater than about 50 G, the entire  $F = 1$  manifold becomes high-field seeking. This process allows us to efficiently pump all of the unwanted  ${}^7\text{Li}$  atoms into an antiguiding mode using a single laser wavelength. Since we do not excite  ${}^6\text{Li}$ , we take a statistical loss of one half of the  ${}^6\text{Li}$  due to the magnetic sublevel projections. Figure 3 shows the radial distributions of the two lithium isotopes entering the magnetic guiding region, as well as their distributions on leaving the tube. The isotope-selective guiding and antiguiding are clearly evident, and the enrichment can be made arbitrarily high by optimizing the geometry of the tube. Once an atom collides with the tube walls, the simulation assumes it sticks.

The results of Fig. 3 were simulated assuming a 0.5-m-long region of quadrupole guiding magnets whose specifications are discussed below. That geometry yields 95% enrichment of  ${}^6\text{Li}$ , and 36.8% of the  ${}^6\text{Li}$  that enters the guiding region survives to be collected. The simulation assumes a beam with a mean velocity of 800 m/s and an initial Gaussian spread of 15 m/s

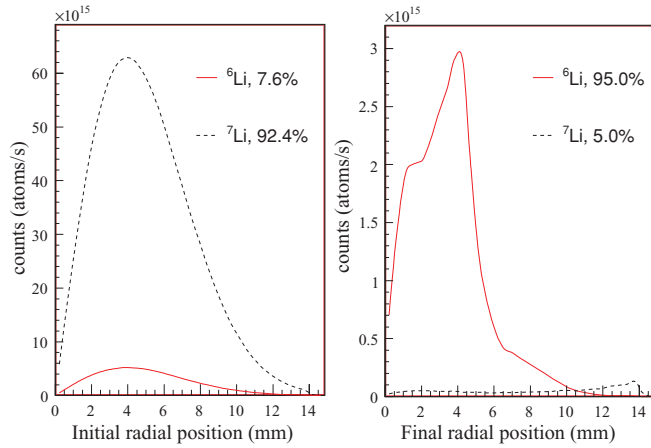


FIG. 3. (Color online) The radial positions of the two lithium isotopes as they enter the magnetic gradient that separates them isotopically, followed by their radial positions on exiting.

in each component of the beam velocity. This velocity corresponds to entraining lithium into a beam of neon. The skimmer shown in Fig. 1 is 5 mm in diameter, and approximately 2% of the beam survives it and enters the multipole tube. Using the previously discussed entrainment and flux estimations, if  $\sim 3 \times 10^{17}$  lithium atoms survive the skimmer, then Fig. 3 implies that we could collect approximately  $10^{16}$   $^6\text{Li}$  atoms/s using this method. Scaling to multiple beams and appropriate pumps would be straightforward.

We simulated a quadrupole magnetic field produced by four permanent magnets for the separation of lithium. The magnets are  $12.7 \times 12.7 \times 25.4$  mm and have a residual flux density of  $B_r = 1.48$  T. The magnets surround a 1.5-cm inner diameter (1.6-cm outer diameter) stainless steel tube. The resulting magnetic flux density, illustrated in Fig. 4, was simulated using finite element analysis. We chose this geometry to avoid putting the magnets in vacuum and to maximize the magnetic field gradients. While a smaller diameter tube will allow for higher field gradients, it will also reduce the total number of collected atoms of the desired isotope. Although our simulations assume the magnets are held in place with an aluminum holder, slightly higher gradients can be achieved by using a carbon steel holder.

One of the distinct advantages of single-photon atomic sorting is that the technique can be applied to almost every atom in the periodic table. Lithium is particularly easy because it has only two isotopes, and its ground state is  $^2S_{1/2}$ . The details of the experimental implementation may vary for different elements. We now discuss two examples that represent qualitatively different categories of elements.

The first example illustrates separation for elements with zero magnetic moment in the ground state and a metastable state that has a nonzero magnetic moment, such as calcium. By using a 272-nm laser, one can excite  $^{44}\text{Ca}$  to the  $^1P_1^o$  state, which quickly decays to the metastable  $^1D_2$  state. Isotopes that are unaffected by the laser will be unaffected by the magnetic gradients since they have zero magnetic moment in the ground state. The low-field seeking state of the  $^{44}\text{Ca}$  will be guided by the magnetic gradients, meaning that a slight bend in the magnetic guiding tube [17] will allow the desired isotope to

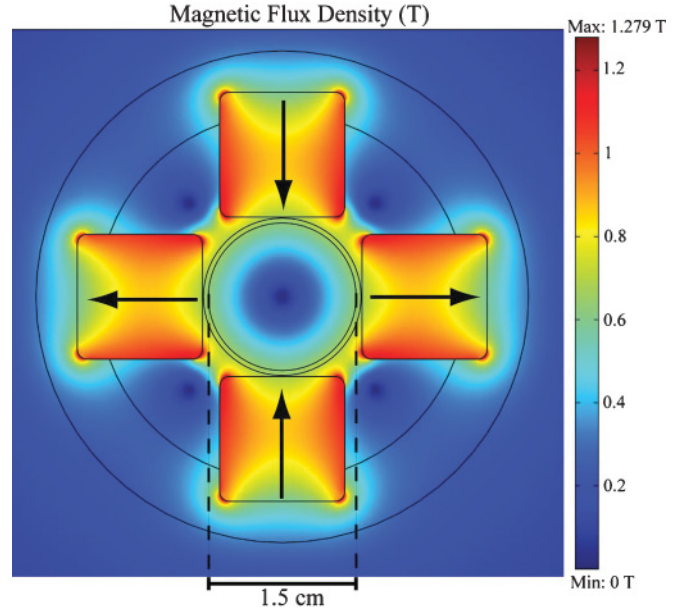


FIG. 4. (Color online) The magnetic flux density of a quadrupole field. The magnets surround a 1.5-cm inner diameter (1.6-cm outer diameter) stainless steel tube and are held in place with an aluminum holder. The magnets are out of vacuum, and the arrows define the direction magnetization.

be collected at high enrichment. Figure 5 shows simulation results of the enrichment of  $^{44}\text{Ca}$ . That simulation utilized a 2-m-long hexapole magnetic field [18] created by six of the same magnets described above only arranged around a larger 2.1-cm inner diameter (2.2-cm outer diameter) stainless steel tube. The tube had a slight bend of 6.0 cm over its 2-m length that served to eliminate the unwanted calcium isotopes. Since calcium is heavier than lithium, it can be entrained into a heavier gas such as argon, which corresponds to a beam with a mean velocity of 500 m/s and a Gaussian spread of 15 m/s in each component of the supersonic beam velocity. As shown in Table I, our simulated setup collected 9.0% of the  $^{44}\text{Ca}$  that survives the skimmer at 99.9% purity.

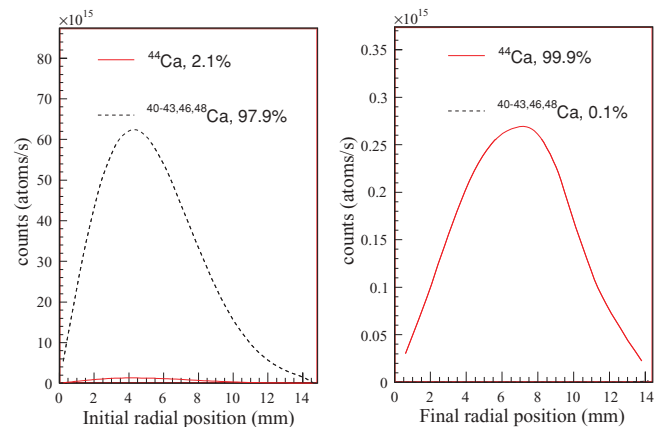


FIG. 5. (Color online) The radial positions of the calcium isotopes as they enter the magnetic gradient that separates them isotopically, followed by their radial positions on exiting.

TABLE I. Simulation results of isotope separation from single-photon atomic sorting.

Target isotope	Natural abundance	Laser $\lambda$ (nm)	Ground state	Guiding length (m)	Enrichment	Collected isotope (%)
${}^6\text{Li}$	7.6%	670.96	${}^2S_{1/2}$	Quad. 0.5	95.0%	36.8
${}^{44}\text{Ca}$	2.1%	272.2	${}^1S_0$	Hex. 2.0	99.9%	9.0
${}^{150}\text{Nd}$	5.6%	471.9	${}^5I_4$	Hex. 2.0	97.9%	23.0

Many elements, however, do not have a suitable metastable state that allows for isotope separation by this method. We propose an alternate implementation of single-photon atomic sorting that is much more general. This method will work on any element that has a nonzero magnetic moment in the ground state, which includes most of the periodic table. As the atoms approach the magnetic multipole guiding, a  $\sigma^-$  polarized laser beam optically pumps our desired isotopes into the stretch low-field seeking state. Simultaneously, a  $\sigma^+$  polarized laser beam optically pumps the other isotopes into the stretch antiguided state. The stretch state refers to the state in which  $m_J$  has the largest possible magnitude. The laser beams can be multipassed through the supersonic beam until almost all of the atoms have been pumped. While relying on optical pumping does mean that more than a single photon has to be scattered on average, the isotope separation is accomplished by the scattering of only a small number of photons, which still makes extremely efficient use of the available laser power. This method does not rely on a long-lived metastable state and is general to all atoms that have a ground-state magnetic moment, although it does typically require multiple laser wavelengths shifted by a few GHz to optically pump all of the isotopes.

We simulate isotope separation using optical pumping for  ${}^{150}\text{Nd}$ , which has a ground state of  ${}^5I_4$ . Using a laser that promotes the ground state to a  $J' = 4$  excited state, such as a 471.9-nm laser [19], Fig. 6 shows we could collect 23% of the  ${}^{150}\text{Nd}$  that survives the skimmer at 98% purity. These simulation results assume a beam with a mean velocity of 500 m/s and a Gaussian spread of 15 m/s in each component

of the supersonic beam velocity. The hexapole magnetic field was a 2-m-long tube like the one described for  ${}^{44}\text{Ca}$  but without the bend. Similar separation results can be achieved using a 1.8-m-long tube with a slight 1-cm bend to aid in eliminating unwanted isotopes. The precise shape of this bend could be further tuned to achieve the optimal separation geometry.

The isotope  ${}^{150}\text{Nd}$  is of particular interest because it is a double  $\beta$  emitter. Many experiments are currently investigating neutrinoless double  $\beta$  decay in order to determine the neutrino mass and whether neutrinos are Dirac or Majorana particles [20]. SNO+ is one such experiment currently under development, and it plans to use a large amount of enriched neodymium to search for neutrinoless double  $\beta$  decay [21]. Enriching neodymium is very difficult and can currently only be done using the atomic vapor laser isotope separation technique [22]. Hopefully this simpler approach can aid in the separation of  ${}^{150}\text{Nd}$ , as well as other isotopes of interest to physics, medicine, and industry.

While the ability to change the magnetic state of a particular isotope and deflect it using a magnetic field is certainly not a new discovery, that capability alone cannot impact the industry of isotope separation. Previous experiments conducted decades ago have demonstrated isotopically selective magnetic deflection [11,23], but the beam fluxes emerging from their sources were typically on the order of  $10^9$  atoms/s, not including factors of isotopic abundance. In the experimental setup we propose, we estimating achieving a flux of more than  $10^{18}$  atoms/s, and rather than being limited to working with atoms in stable gaseous compounds, we can apply our separation technique to almost every element in the periodic table.

In conclusion, we have presented single-photon atomic sorting as a very general and scalable approach to isotope separation. One of the advantages of scaling the technique is that the laser can be recycled in a multipass configuration until it is depleted. For example, a laser with 1 W of power can deliver approximately  $10^{19}$  photons/s, which means that even such a modest laser could be used to separate the output from multiple supersonic nozzles in a mass-production setting. A supersonic beam can be operated in a continuous or quasicontinuous mode, and the flux is limited only by available vacuum pump speed. Large-scale separation seems feasible using either diffusion pumps or cryopumps. The next step will be a first experimental demonstration of single-photon atomic sorting.

M.G.R. acknowledges support from the State of Texas Advanced Research Program and the Sid W. Richardson Foundation.

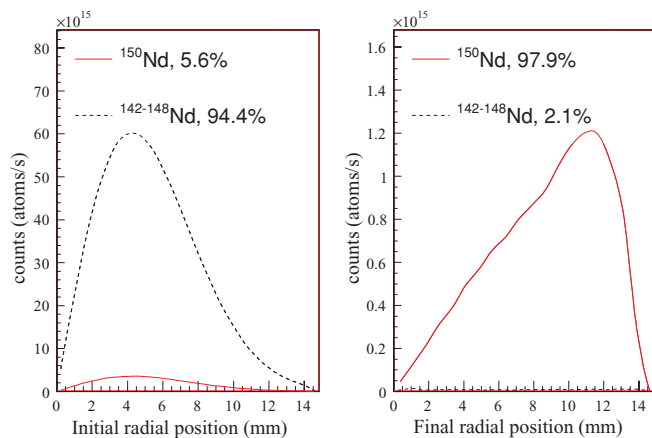


FIG. 6. (Color online) The radial positions of the neodymium isotopes as they enter the magnetic gradient that separates them isotopically, followed by their radial positions upon exiting.

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