

Counting the atoms

Practically every element in the periodic table can be detected, down to single-atom sensitivity, by resonance-ionization-spectroscopy methods involving commercially available lasers.

G. S. Hurst, M. G. Payne, S. D. Kramer and C. H. Chen

The title of this article was inspired by a similar one used by Ernest Rutherford for an unpublished lecture note now in the University of Cambridge library. Rutherford's title was "The Counting of Atoms," by which he could have meant the counting of individual ionized helium atoms (alpha particles), or, alternatively, he may have had in mind "decay counting" as an indirect indication that a parent atom had transmuted. With the modern pulsed laser, Rutherford's idea becomes entirely practicable, and individual atoms can now be counted by several different methods, the subject of the present article. Our group at Oak Ridge has demonstrated these methods with apparatus like that shown in figure 1.

Let us examine more carefully what we mean by "counting the atoms." Figure 2 shows, in tabular form, four different ways of going about it. The first, "decay counting" (called method a in the figure) is an observation of the decay of a parent atom; this is the method Rutherford used when he detected alpha decay from radon. With the laser techniques to be described below, the parent atoms can be counted directly—whether they are stable or unstable. This is method b in the figure. Our group at Oak Ridge has also shown recently that the daughter atoms can be counted in a time-coincident observation with the decay. We call this "decay-daughter counting," and illustrate it as method c in figure 2. And finally, a technique currently under development should allow direct counting of daughter atoms after they have accumulated on a surface or in a gas, thus giving a direct measurement of the number of parent atoms that have decayed during a protracted time period. This is "direct-daughter counting," method d in figure 2. We should

make it clear at this stage that the "counting of atoms" requires all of the following:

- ▶ it must be possible to recognize the type of atom in question unambiguously without interference from atoms or molecules of another type;
- ▶ the sensitivity must be great enough to detect even one atom; and
- ▶ a system of memory is required so

that an atom, once sorted, is not detected more than once.

Because of these restrictions, we are not discussing the very sensitive laser fluorescence techniques,¹ which usually require time averaging over many atoms to obtain a signal and do not normally provide a means for sorting the atoms. We shall, therefore, limit the discussion to the use of pulsed



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lasers that can be used to remove one electron from each atom in a population of atoms of a selected type, through a variety of photo-ionization schemes we call "resonance ionization spectroscopy." Once an atom is ionized it can be directed into detectors and stored there for almost any desired time.

Resonance ionization spectroscopy

Since 1949 it has been clear that thermal electrons can be individually counted. S. C. Curran, A. L. Cockcroft and J. Angus² showed in that year that a gas proportional counter can detect electrons with nearly unit efficiency. Now a variety of electron multipliers is available for the same purpose. So the problem of counting atoms is solved if we can ionize every atom in a population and then count the free electrons.

Energetic charged particles and x rays both strip electrons from all types of atoms and molecules in their path, but the ionization process is not selective and the efficiency of ionization is low. For example, an alpha particle moving down a column of helium 1 mm in diameter until the particle comes to rest will ionize only one atom per 10^{13} atoms present. One of the many re-

markable features of the laser is that it can be used to ionize *all* atoms of a given type and essentially *none* of another type. Thus, even in larger volumes (say, 1 cm in diameter and many centimeters in length) one atom of a given type can be ionized with a pulsed laser that does not ionize any of the extremely large number of atoms of another type that may be present. This feature is the key to counting atoms by resonance ionization spectroscopy.

In a recent review article³ members of our group showed that tunable pulsed lasers can accomplish the removal of one electron from each spectroscopically selected atom, and that this is possible by several different processes. Incidentally, we called this method "resonance ionization spectroscopy"^{4,5} to distinguish it from the non-selective ionization associated with x rays and radioactivity. Our first use of the method, oddly enough, brought us into contact with both selective and non-selective ionization (see figure 3). We selectively ionized metastable helium atoms that had been excited by charged-particle beams to the 2^1S state; the exciting beam simultaneously produced copious non-selective ionization.

The five different methods of carrying out resonance ionization spectroscopy are illustrated in figure 4. It was the first and most elementary of these that we used to detect 2^1S helium. Ground-state atoms of nearly all the known elements are now amenable to the method using one or other of the five schemes of figure 4 and employing only commercially available tunable lasers.

The volume that can be saturated during one laser pulse depends critically on the type of atom. To see why this is so, refer to scheme 1 in figure 4. When a laser is pulsed into a region of space, each atom in that space can be rapidly excited to a bound level provided the photons are of the correct frequency, ω_1 , that is, when $\hbar\omega_1$ is the energy difference between the initial and final states. In fact, at a resonance, atoms are excited and de-excited by stimulated emission so rapidly that a quasi-equilibrium between excited states and ground states is established very early (say within 10^{-11} sec) in a laser pulse that might be of 10^{-6} -sec duration. The expression "quasiequilibrium" is used because the excited-state population is photoionized to continuum states by photons with frequency ω_1 at a lower rate. Even the rate of the slower photoionization step can be

great enough that at the end of the laser pulse essentially all of the atoms will have been ionized. Under this condition, one electron is removed from each atom of a spectroscopically selected type, and the resonance-ionization-spectroscopy process is saturated. But saturation requires that all excited atoms will be ionized with the slower continuum process. Because cross sections for photoionization of excited states are typically 10^{-17} cm², more than 3×10^{17} photons per cm² must be delivered in one laser pulse in order to detect more than 95% of the atoms. In the visible region, therefore, an energy density of about 100 mJ/cm² is required for saturation. Flashlamp lasers can deliver about one joule per pulse; hence, the beam area can be 10 cm² and the length of the detector volume can be as long as the wire of the ionization detector, which can be a proportional counter or a Geiger-Müller tube. Sensitive volumes in scheme 1 of figure 4 could easily be 100 cm³ if needed. In scheme 2, where the laser must be frequency doubled to reach the first one-photon allowed level of the atom, volumes can also be large because a large fraction of the fundamental radiation is still in the laser beam. Frequency doubling generates a weak beam for exciting a highly allowed transition, and a strong fundamental beam remains for the more difficult continuum ionization. Only in scheme 5 (needed for atoms such as hydrogen, krypton and xenon) must the saturated volume be rather small. Two-photon excitation from the ground state to levels of the same parity is a low-cross-section process. However, with recently available excimer lasers in the far ultraviolet, volumes of 0.1 cm³ or greater can be saturated even with scheme 5.

The table on page 27 shows the applicable schemes for resonance ionization spectroscopy of ground-state atoms of all the known elements except helium and neon.

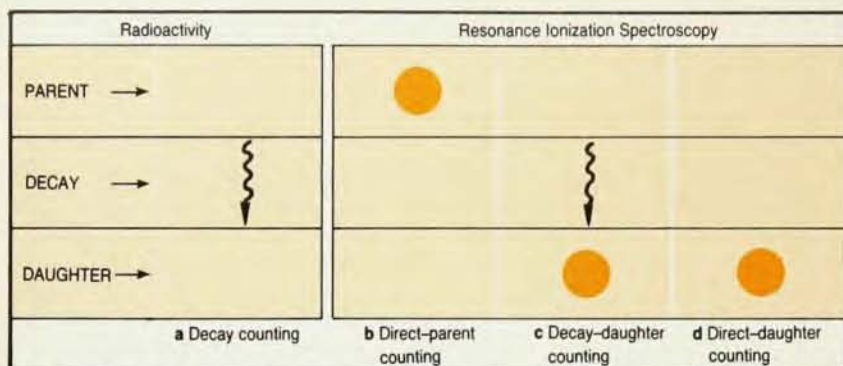
Stable atom counting

The techniques we have discussed so far represent the "spectroscopy" aspect of resonance ionization spectroscopy—that is, detection and identification of specified atoms. The possibility of direct counting of stable atoms follows immediately if a proportional counter with one-electron sensitivity is used. The patent on this technique dates from 1974 (filing date) and 1976 (granted); it was issued to Sam Hurst, Marvin Payne and Bryan Wagner of this Oak Ridge group.⁶ The first demonstration,⁷ by Hurst, Munir Nayfeh and Jack Young followed a year later in 1977.

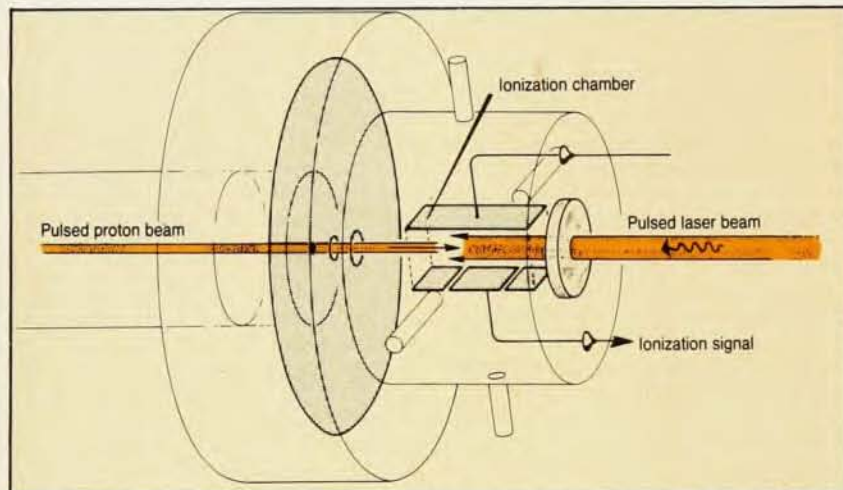
In figure 5 we illustrate in a highly schematic fashion the first demonstration of one-atom detection. A laser



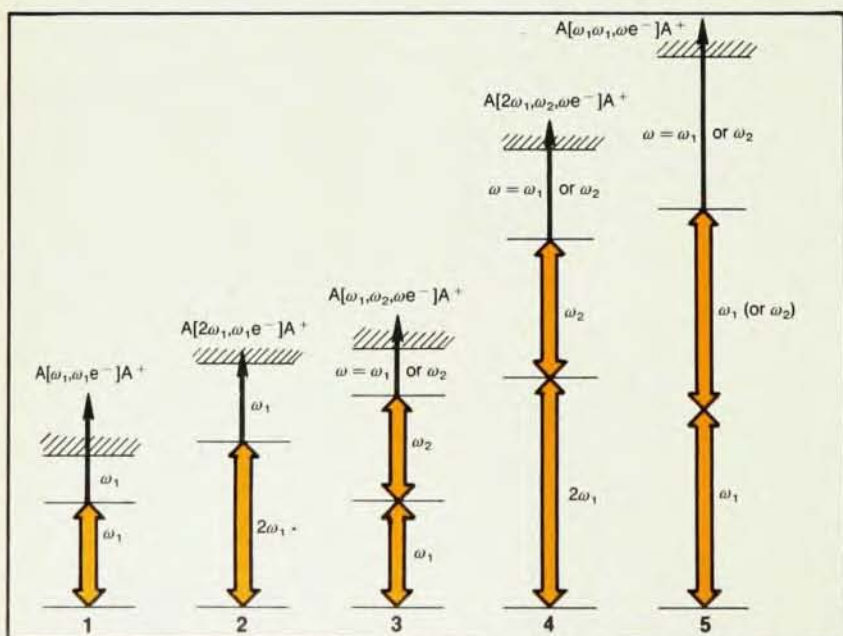
Resonance-ionization spectroscopy apparatus. Greg Foltz (right), an Oak Ridge postdoctoral fellow, holds a combined drift chamber and proportional counter used in conjunction with the pulsed lasers seen in the background. Steve Allman (left) makes an adjustment. (ORNL photo) Figure 1



Four different ways of counting atoms. The column on the left shows, schematically, the radioactive-decay method familiar from the work of Rutherford. The other three columns show the three types of resonance-ionization-spectroscopy discussed in this article. Figure 2



The first demonstration of resonance-ionization spectroscopy used this device to obtain the absolute population of $\text{He}(2^1\text{S})$ created by charged-particle excitation of helium. A large-area pulsed laser beam selectively ionized all the $\text{He}(2^1\text{S})$ atoms remaining in the beam. Figure 3



With these five laser schemes all the known elements except helium and neon can be ionized. The schemes are classified here by a notation similar to that used in nuclear physics. Figure 4

beam is directed through a proportional counter with wavelength and intensity specifications such that any atom of a chosen type in the volume probed by the pulsed beam is resonantly ionized. With Curran, Cockroft and Angus's improved Rutherford-Geiger counter,² a single electron from any one atom that might be in the laser volume can be amplified sufficiently that a pulse can be recorded with conventional nuclear counting equipment. In the experiment,^{7,8} just one atom of cesium in a background of 10^{19} atoms of argon and 10^{18} molecules of CH_4 was easily detected. Time and space resolution are inherent in the method because the laser is pulsed into a defined region of space.

This proportional-counter method of one-atom detection has been used in a wide variety of applications. This variety is based, in part, on the work of Larry Grossman and other members of our group,^{9,10} who showed that it is advantageous to work with alkali-halide molecules (for example CsI) and to make use of two pulsed lasers, one to liberate atoms (in the CsI example, cesium) at $t = 0$, and the other to detect the free atoms at $t > 0$. In this way the cross section for photodissociation of an alkali halide could be obtained in absolute units without knowledge of the vapor pressure of the molecule.⁹ Then, time-delay studies¹⁰ demonstrated that both chemical reaction rates and diffusion rates of free atoms in a reactive environment could be obtained. This work established a very convenient way to study small but well-controlled populations of a few atoms. For instance, a quantity in radiation physics known as the Fano factor can be measured by observing the fluctuations of pulse height in a proportional counter due to a low-energy x-ray source, say 5 keV, compared with the fluctuations of free atom concentration in a gas.¹¹

An extension of the technique in which drift chambers and proportional counters are both used in combination with pulsed lasers has evolved into a versatile apparatus (see figure 1), which has also been used to make precision measurements of the diffusion of free atoms, enabling the testing of diffusion theory in spatial and temporal domains.¹² Analytical applications of the resonance-ionization-spectroscopy proportional-counter method have been made, for instance, by Santos Mayo and Thomas Lucatorto at the National Bureau of Standards to measure sodium atoms in high purity silicon materials and by Andrzej Miziolek who has developed a sensitive mercury detector at the Scripps Institution of Oceanography.

The list of applications of one-atom detection using proportional counters is still increasing. For instance, Ja-

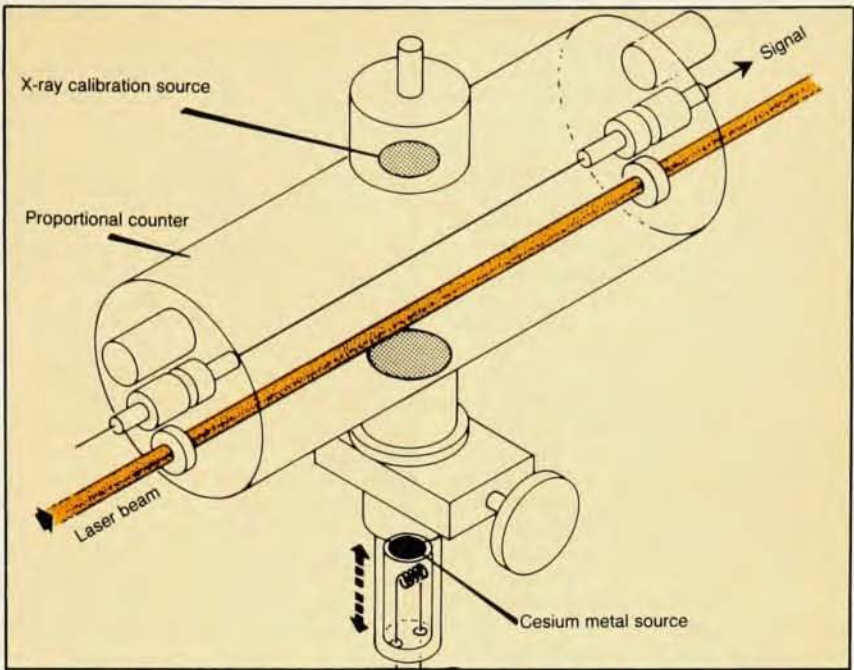
cinto Iturbe (visiting our group from the Universidad del Pais Vasco, in Bilbao, Spain) is undertaking three new applications: the systematic study of the statistics of various physical phenomena such as atom-atom collisions; an experimental test of the ergodic hypothesis for freely diffusing atoms, and autocorrelation of free-atom concentrations. The studies of autocorrelation will be made with the help of a three-dimensional position-sensitive proportional counter designed by M. K. Kopp of Oak Ridge's Instrumentation and Controls Division. With this arrangement, we expect also to be able to characterize the beam quality of a pulsed laser in three-dimensional detail and to see the evolution of diffusive clouds of free atoms.

Prompt daughter-atom counting

A very basic extension of the resonance-ionization-spectroscopy method for counting atoms was the demonstration¹³ that a single atom of cesium could be detected in time coincidence with the fission decay of a single Cf^{252} nucleus. This experiment has significance in charge-exchange research because it showed that the Cs^{n+} ion born from the nucleus of another atom could become neutral Cs^0 in the process of losing its fission recoil energy in a gas. But perhaps greater significance lies in the more general implication that daughter atoms can be detected in time coincidence with the decay of the parent, as suggested in part c of figure 2.

In a collaboration with Ray Davis and his group at Brookhaven National Laboratory, we seek to count the small number of atoms generated in massive targets by prolonged exposure to neutrinos from the Sun. First, let us emphasize that Davis's group has already successfully accomplished one such measurement using nuclear counting methods¹⁴ (decay counting, part a of figure 2). In their work, small numbers of Ar^{37} atoms created by neutrino interaction with Cl^{37} were detected by placing the sample of Ar^{37} in a small proportional counter. Anticoincidence shields, rise-time discrimination, and many other precautions resulted in backgrounds so low that Ar^{37} production rates of one atom per day could be measured.

However, the Brookhaven group and John Bahcall¹⁵ (Institute for Advanced Studies, Princeton) are also interested in neutrino detectors such as gallium and lithium, because the neutrino flux, as measured by chlorine with a threshold at 814 keV, is much lower than expected from the standard solar model. Detection of Be^7 from the lithium reaction $Li^7(\nu,e^-)Be^7$ for neutrino energies greater than 861 keV would be a nice example of the use of one-atom detection, as illustrated in part c of



Single-atom detector. In this 1977 experiment any atom of a chosen type in the volume probed by the laser beam could be ionized and counted by the proportional counter. Figure 5

Applicability of Resonance Ionization Spectroscopy schemes

Element	Atomic number	Scheme	Element	Atomic number	Scheme	Element	Atomic number	Scheme
Hydrogen	1	5	Bromine	35	5	Thulium	69	1
Helium	2	-	Krypton	36	5	Ytterbium	70	2
Lithium	3	2	Rubidium	37	1	Lutetium	71	1
Beryllium	4	4	Strontium	38	2	Hafnium	72	3?
Boron	5	4	Yttrium	39	3	Tantalum	73	3
Carbon	6	5	Zirconium	40	3	Tungsten	74	3
Nitrogen	7	5	Niobium	41	2	Rhenium	75	2
Oxygen	8	5	Molybdenum	42	2	Osmium	76	3
Fluorine	9	5	Technetium	43	3	Iridium	77	3
Neon	10	-	Ruthenium	44	2	Platinum	78	4
Sodium	11	2	Rhodium	45	2	Gold	79	4
Magnesium	12	3	Palladium	46	4	Mercury	80	4
Aluminum	13	1	Silver	47	4	Thallium	81	1
Silicon	14	4	Cadmium	48	4	Lead	82	4
Phosphorus	15	5	Indium	49	1	Bismuth	83	4
Sulfur	16	5	Tin	50	2	Polonium	84	4
Chlorine	17	5	Antimony	51	4	Astatine	85	5?
Argon	18	5	Tellurium	52	4	Radon	86	5
Potassium	19	1	Iodine	53	5	Francium	87	1?
Calcium	20	2	Xenon	54	5	Radium	88	2
Scandium	21	2	Cesium	55	1	Actinium	89	3?
Titanium	22	2	Barium	56	2	Thorium	90	1?
Vanadium	23	2	Lanthanum	57	1	Protactinium	91	1?
Chromium	24	2	Cerium	58	1	Uranium	92	2
Manganese	25	2	Praseodymium	59	1	Neptunium	93	1
Iron	26	2	Neodymium	60	1	Plutonium	94	1
Cobalt	27	2	Promethium	61	1?	Americium	95	2
Nickel	28	2	Samarium	62	1	Curium	96	1
Copper	29	4	Europium	63	2	Berkelium	97	1
Zinc	30	4	Gadolinium	64	1	Californium	98	1?
Gallium	31	1	Terbium	65	1	Einsteinium	99	2
Germanium	32	4	Dysprosium	66	1	Fermium	100	1?
Arsenic	33	5	Holmium	67	1	Mendelevium	101	1?
Selenium	34	5	Erbium	68	1	Nobelium	102	1?
						Lawrencium	103	1?

The schemes referred to here are those of figure 4, above

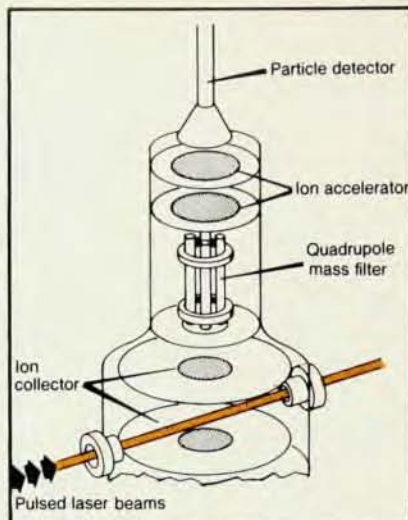
figure 2. The decay of Be^7 back to Li^7 by electron capture is accompanied by an Auger electron which, unfortunately, can only give a faint and erratic radiation signature in a detector. However, a pulse derived from a proportional counter can be used to fire a laser each time a low-energy electron triggers the counter. And if the laser is tuned to ionize the lithium daughter atom, the detection of a daughter in time-coincidence with the Auger electron should be a good indication of the decay of one atom of Be^7 .

Such a system is under active development¹⁶ for the lithium solar-neutrino experiment, and it has a number of other possible uses. For example, it could be used to detect Ar^{39} removed from ocean water. This isotope, created in the atmosphere by cosmic rays, serves as a steady-state source for the study of oceanic mixing processes; its 270-year half-life and chemical inertness make it well suited for the purpose. We are collaborating with Harmon Craig and R. D. Willis of the Scripps Institution of Oceanography to develop a sensitive Ar^{39} detector.

The main obstacle in detecting the lithium daughter atom (or daughter atoms in general) may be that the daughter is thermalized as Li^+ , which must be neutralized before we can detect it by resonance ionization spectroscopy. The solution to this problem could, however, lead us to "resonance ionization spectroscopy with amplification," which has additional advantages.¹⁷ The basic idea is that positive ions can be rapidly neutralized in photo-enhanced collisions with molecules of low ionization potentials, yielding atoms that can be reionized with resonance ionization spectroscopy, thus completing one cycle. If these neutralization-ionization cycles can be completed in a time less than the length of a laser pulse (one microsecond), then amplification of the ionization signal is possible. One may view the whole process as a cycle that can be initiated in either the ground state or the ionization continuum of an atom; hence, regardless of whether the daughter atom begins its life as a neutral or a positive ion, it can be detected. And the gain in the signal comes as a bonus!

Maxwell's sorting demon

In 1871, James Clerk Maxwell visualized a demon that could see atoms and would have sufficient intelligence to perceive the velocity of an atom (or, alternatively, the type of atom). On the basis of this information, the demon could open and close doors to accomplish the "sorting of atoms." Such a sorting demon would violate the second law of thermodynamics and was thus a major conceptual problem in physics until Louis Brillouin, in 1951,



Maxwell's sorting demon in a version soon to be tested at Oak Ridge. Single atoms will be simultaneously counted and stored on the detector surface—which may be an electron multiplier. Figure 6

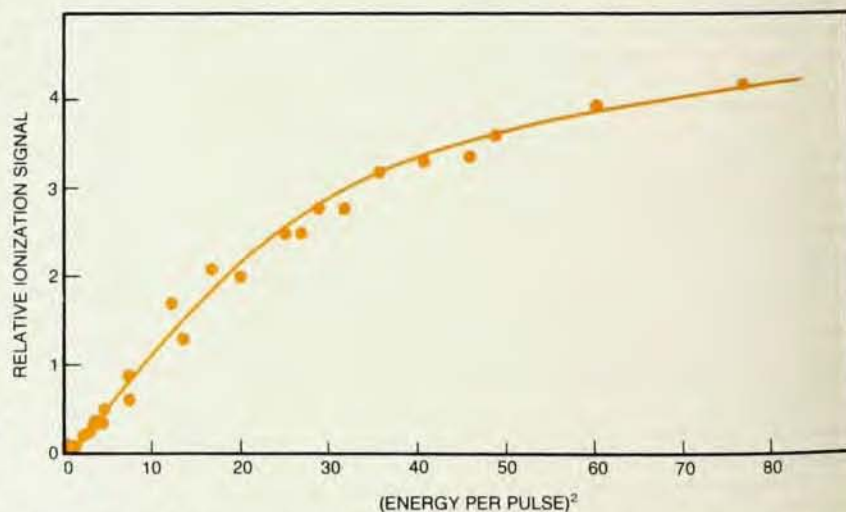
showed that the demon would have to be equipped with a flashlight to see the atom. Taking into account the entropy increase in the flashlight, the conceptual problem was removed; yet the idea of a prankish demon remained intact. We are designing experiments that will constitute a new form of one-atom detection through practical realization of the Maxwell demon.

Samples containing ten or more atoms of argon, krypton or xenon will be subjected to our version of Maxwell's sorting demon, whose internal machinery is shown in figure 6. Pulsed laser beams are tuned to produce the resonance-ionization-spectroscopy process in, for example, xenon. After ionization, Xe^+ ions are accelerated to about

10 keV and implanted into a detector surface such as a channel electron multiplier. Thus a single atom may be simultaneously counted and stored! With implantation depths on the order of 100 Å, atoms may be kept in the solid for hours before being lost by diffusion. Thus, the demon has hours in which to count and store on the order of ten atoms—a feat requiring only limited intelligence.

A more challenging problem for the sorting demon is to count a few inert atoms of a given isotope amid a background of many more atoms of another isotopic species. For this, the intellectual prowess of the sorting demon must be enhanced by providing him with a small mass spectrometer, judiciously placed between the laser beam and the ion counter. In this way, only those isotopes of interest are accelerated and implanted into the ion counter. However, a small mass spectrometer will have limited resolution, so that the rejection ratio between adjacent mass peaks may be limited to, say, 1000. In the cases where much larger isotopic ratios must be dealt with, we visualize that the atoms would be returned to the cell in a short time by heating the detector slightly. This cycle can now be repeated n times to achieve, in our example, the detection of one atom of the desired isotope out of 1000ⁿ atoms of other isotopes.

The practical realization of Maxwell's sorting demon has recently been achieved¹⁸ as described (essentially) in an earlier patent application.¹⁹ Figure 7 shows the ionization signal versus the square of the laser energy per pulse when xenon was detected in the process described as scheme 5 in figure 4. This curve shows that even with a noble gas such as xenon, commercial Nd:Yag laser systems can ionize nearly all of the



Ionization signal as a function of the square of laser energy per pulse, for the detection of xenon by process 5 of figure 4. Energy per pulse is about 1 mJ at the maximum, and the beam from a commercial Nd:YAG laser system is focussed to a waist of 0.2 mm. Figure 7

atoms in a small volume. But it is not essential that saturation of the ionization process be achieved, for the nature of the demon is to keep looking until all of the atoms have been detected and sorted into another compartment. Demonstration of the storage of the atoms in the detector is shown in figure 8. In the Nd:Yag laser system used the effective volume that could be cleared of each atom was only $2 \times 10^{-4} \text{ cm}^3$, consistent with the long running time shown in figure 8. However, with modern excimer lasers, the effective volume can readily be increased to $5 \times 10^{-3} \text{ cm}^3$ and the lasers can be operated at rates of 100 Hz or more. Thus volumes of a few liters (large enough to accommodate quadrupole mass filters) can be cleared of nearly all the atoms in about one hour. Consequently, it is now possible to operate Maxwell's sorting demon, with isotopic selectivity at quite satisfactory rates when counting atoms in the laboratory.

A solar-neutrino experiment using the reaction $\text{Br}^{81}(\nu, e^-) \text{Kr}^{81}$ could require detecting 500 atoms of Kr^{81} in 10^9 atoms of krypton from atmospheric contamination in the neutrino target tank. On the other hand, Kr^{81} detection for the dating of polar ice requires counting 1000 atoms of Kr^{81} (from only one liter of ice) out of 1.4×10^{15} Kr atoms.²⁰ Another application of Kr^{81} is for geochronology, because its lifetime of 2.1×10^5 years provides a convenient time scale for geological events.²¹ For oceanic circulation the problem is the detection of Ar^{39} in the ocean—which contains 6000 atoms of Ar^{39} and 10^{19} atoms of argon per liter of ocean water at the surface. Since Ar^{39} is created in the atmosphere above the ocean and is automatically swept into the surface water, it can be sampled as a function of depth to profile oceanic turnover rates. The half-life of Ar^{39} is 270 years and is ideally on scale with the crudely estimated elapsed time since surface exposure of deep Pacific and Atlantic waters (1000 years at the greatest depth).

One application of the xenon detector could be to indicate levels of plutonium and other fissionable materials in radioactive wastes. Our work on this application was done in collaboration with M. R. Cates (Los Alamos) and with H. M. Borella and L. A. Franks (EG & G). In this concept, barrels of potentially contaminated materials would be irradiated with neutrons (or photons) to produce fission in which xenon and krypton are prominent fission products. After irradiation, even a few xenon atoms can be recovered and counted by the Maxwell sorting demon. Furthermore, the demon we have described can serve as a selective pump to clear atoms or isotopes of a particular type from a region of space.

We have described above techniques for direct counting of parent atoms (part b of figure 2), decay-daughter atom coincidence counting (part c of figure 2), and the application of both of these to Ar^{39} counting for oceanographic research. We can also illustrate our case of delayed daughter counting (part d of figure 2) with Ar^{39} . This technique is being pursued by J. E. Parks (Western Kentucky University), E. T. Arakawa (Oak Ridge) and R. D. Willis (Scripps). The task here is to sort out daughter atoms that have accumulated on a surface. When Ar^{39} atoms beta decay, K^+ ions are produced as daughter atoms. These ions could be collected on a negatively charged surface for about six months. In this relatively long time period about 100 of the Ar^{39} atoms would decay into K daughter atoms on a known surface. To prevent formation of unknown chemical states of potassium, it may be prudent to form KI on a surface previously coated with NaI or impregnated with HI. Detection of the few KI molecules would involve the following sequence: First KI is photodesorbed and then photodissociated; potassium is subsequently ionized, accelerated and implanted into a surface detector. This is another example of Maxwell's sorting demon, for here he must count only one molecule at a time, and continue to count until the vessel is devoid of the type of molecule that the demon is trained to catch.

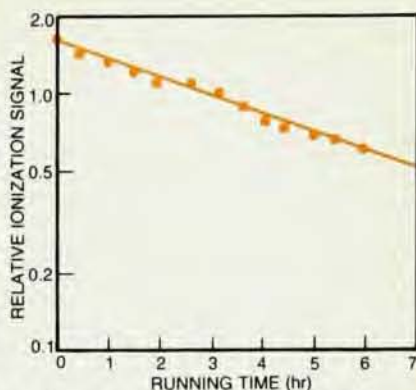
In the past few years we have learned how to "count the atoms," that is, to actually enumerate all the atoms of a given type in a sample. And while we do not know exactly what Rutherford had in mind by "The Counting of Atoms," he may well have had the essential ideas of this article. If so, he lacked only the laser and the perfection of his own detectors (the "Rutherford-Geiger" counter) to realize his ideas.

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This work was supported primarily by the US Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation, and in part by the Office of Naval Research.

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Depletion of xenon atoms in the cell for the same conditions as figure 7. The effective volume that could be cleared of specific atoms was only $2 \times 10^{-4} \text{ cm}^3$, thus yielding the long clearance time shown here. More powerful lasers with larger effective volumes would reduce the time considerably. Figure 8

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