Mercury Isotopes for Efficient UV Lamps and Fluorescent Lighting

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Abstract—Mercury-discharge lamp technology is the basis for fluorescent lighting and UV lamps for water purification. We showed that by modifying the ratios of the seven stable isotopes of mercury from their natural relative abundances, lamp efficiencies can be improved by over 20%. In parallel, recent methods of isotope separation can make the cost of the optimized isotopic mixtures economically competitive. We review these recent developments and discuss implications for future lighting and UV lamp technologies.

Index Terms—Fluorescent lamps, isotope separation, isotopes, ultraviolet generation, water pollution.

I. INTRODUCTION

ERCURY-DISCHARGE fluorescent lamps have dominated the lighting market for many years. Despite the rapid growth of the LED market, it is likely that linear fluorescent lamps will continue to persist for several decades due to the many billions of fixtures installed worldwide. Development of new phosphors that do not require rare earths was reported recently in a press release by the Department of Energy, indicating an ongoing investment in fluorescent lighting technology [1]. Other advances include a steady decrease in the amount of mercury used in each lamp, enabled by more precise methods of dispensing as well as improvements in materials to reduce the absorption of mercury inside the lamp. Further improvements in energy efficiency and lamp lifetime are incremental but have been steady over the years. Recently, we proposed a method for dramatically improving the energy efficiency of fluorescent lighting, simply by using an optimized mixture of mercury isotopes instead of natural mercury [2]. This is a dropin replacement, which requires no redesign of lamps, that could be readily adopted for use.

Meanwhile, a rapidly growing market is UV water purification, where mercury lamps offer the best current technology and there is no solid-state alternative at this time. The main method of water purification today uses chlorine, but there are increasing concerns about the efficacy of this method with the emergence of chlorine-resistant pathogens, such as cryptosporidium, E. coli,

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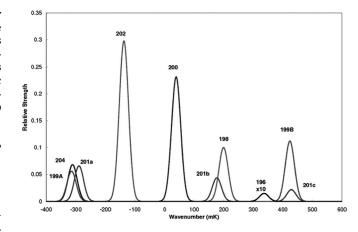


Fig. 1. Spectrum at 254 nm of the seven stable isotopes of Hg. The signal size represents the natural abundance of each isotope as follows: Hg-196 (0.146%), Hg-198 (10.02%), Hg-199 (16.84%), Hg-200 (23.13%), Hg-201 (13.22%), Hg-202 (29.8%), and Hg-204 (6.85%). Only the odd isotopes of Hg (199 201) have hyperfine structure, and these lines are denoted by a and b, respectively.

legionella, norovirus, and shigella [3], [4]. In addition, chlorine creates dangerous trihalomethanes, such as chloroform, as byproducts in the water [5]. These serious concerns have accelerated the adoption of UV water purification, which uses UV light at a wavelength of 254 nm from mercury lamps and is highly effective at destroying any pathogens. The water is filtered prior to passing through the UV lamps, and the exposure level ensures that no pathogens can survive. In addition, this method does not produce any toxins or carcinogens. Major cities are now adopting UV purification systems, such as New York and Chicago, and most municipalities throughout the world are likely to follow [6].

II. MERCURY ISOTOPES IN DISCHARGE LAMPS

During the operation of a mercury gas discharge lamp, mercury atoms are excited and emit radiation at several wavelengths including 254 nm. The high opacity of the gas causes the 254-nm photons to undergo multiple scattering before exiting the lamp. Collisions between atoms can transfer the excitation to a nonradiating state, which results in a loss of efficiency. It was proven experimentally that changing the percentage of a particular isotope (Hg-196) led to a 7% improvement in UV efficiency [7]. This effect is due to the reduction of the opacity of the gas, since the isotope shift in mercury is much larger than the Doppler width. A spectrum of the seven stable isotopes of Hg is shown in Fig. 1.

In recent work by Lawler and Raizen, it was shown that by tailoring the percentage of the seven stable isotopes of mercury, an improvement of up to 22% in efficiency can be obtained by

reducing the opacity of the gas, thereby increasing the escape rate of the 254-nm photons [2]. The paper presents a numerical study using a Monte Carlo code that has been well tested in the past with respect to actual lamps. This composition was awarded a U.S. patent in less than one year after filing [8]. This change would be a drop-in replacement, requiring no modification in the lamp design, just a different ballast resistor. A lower electrical consumption for the same UV light output would also likely lead to longer lamp lifetime, although this effect must be tested. This leads to the next question: What is the cost of isotope separation for mercury, and its scalability to industrial quantities?

III. SEPARATION OF MERCURY ISOTOPES

Prior methods of separation, such as the calutron and photochemical separation, are prohibitively expensive and therefore impractical. The calutron is a method of electromagnetic separation, invented by Ernest Lawrence in 1930, and originally applied to uranium separation [9]. After World War II, the large calutron facility at Oak Ridge became a general-purpose machine that could provide enriched isotopes for most elements in the periodic table. It was shut down permanently in the late 1990s, and the only large-scale calutron operating today is in Russia [10]. Despite years of effort to use lasers to directly ionize isotopes, it did not prove to be a cost-effective or scalable approach [11]. The photochemical method, specifically applied to Hg isotopes, is expensive and poses a serious safety hazard involving flammable gases and a highly toxic mercury compound.

In the past few years, the Raizen group first proposed and then demonstrated experimentally a new and highly efficient method for isotope separation [12], [13]. The result, published in *Nature Physics*, shows that the energy efficiency is several orders of magnitude better than the calutron [13]. Magnetically activated and guided isotope separation (MAGIS) relies on lasers or other tunable light sources to optically pump the desired isotope into a particular magnetic state that is repelled by magnetic fields, followed by a slightly curved array of permanent magnets. The experimental demonstration was with atomic lithium, which has two stable isotopes: Li-6 (natural abundance of 92.4%) and Li-6 (natural abundance of 7.4%). The goal was to deplete the Li-6 content, which is needed for cooling water in nuclear reactors. This work showed that in a single-pass, Li-7 was produced with a purity of over 99.95%, which is the standard of the nuclear industry.

MAGIS is ideally suited to the separation of mercury isotopes, facilitated by the zero magnetic moment ground state and the near-room temperature operation. In addition, optical pumping can be accomplished with mercury lamps that use the desired tailored isotopic composition, which is a simpler and more scalable approach than lasers. The only concern is the toxicity of mercury, but this can be safely contained. A patent on the MAGIS process was also awarded in less than one year after filing [14]. A more detailed description of the MAGIS process applied to mercury isotopes is now described.

The method has three main steps:

1) the source, which generates the stream of mercury atoms;

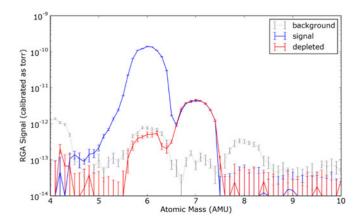


Fig. 2. Mass spectrum of Li with and without the optical pumping beam. In the experiment, enriched Li-6 was used in order to improve the dynamic range. The blue curve is without optical pumping. The red curve is with the optical pumping and shows depletion of the Li-6 component without affecting Li-7.

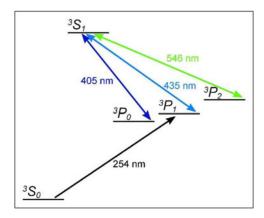


Fig. 3. Simplified Grotrian diagram of neutral Hg. The energy levels are not shown to scale.

- 2) the state preparation, which prepares the stream of atoms into isotopically selected states that allows magnetic separation;
- the guiding and collection, which separates the prepared atoms into trajectories that have modified isotopic abundances.

All of this will take place in a large high-vacuum chamber maintained at a background pressure low enough to ensure that collisions with background gas will not deflect the target atoms from their desired trajectories through the apparatus. This pressure should be below 10^{-4} Pa, which can be easily achieved by state-of-the-art vacuum pumps. This method can be scaled up in parallel with multiple chambers. Atoms that do not enter the guides can be collected on a surface for reuse. Specific to mercury, due to the fact that it is in liquid state near room temperature, a closed flowing system can be easily implemented.

A. Source

The starting point is to create a flowing stream of neutral Hg atoms. A source can be refilled with the pure element (the feed-stock) without having to open the vacuum chamber by flowing liquid Hg through a system of valves. The oven chamber will be constructed from stainless steel and follow an existing

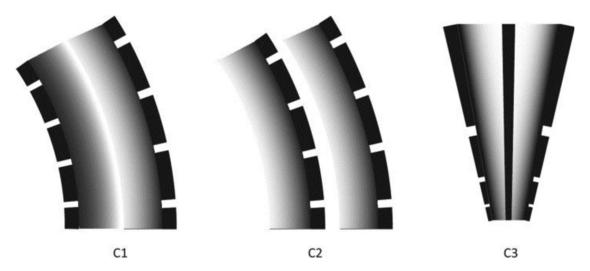


Fig. 4. Depiction of three planar arrays for magnetic separation. The thick black-dashed lines represent the magnets with the magnetic field strength proportional to the darkness of the shading in the guiding region. The solid black line in C3 represents a nonmagnetic collection plate at the field minimum.

design that produces a collimated beam with a desired angle of divergence. The principle is to rely on an array of capillary tubes where the ratio of diameter to length determines the angle of divergence. Such an oven has been demonstrated in atomic lithium [15] and should work well for mercury. The effective area can be scaled up to produce a brighter atomic beam without changing the beam divergence. The limitations imposed by the source itself will just be the maximum pressure one can achieve without cluster formation, and any temperature-related constraints. While there is the possibility of collisions of the atoms with each other, the rate does not become significant until even higher densities, and only a fraction of these collisions will be detrimental.

B. Isotopic State Preparation

The next step is to excite the desired atoms to an appropriate internal state, a process known as optical pumping. The ground state of mercury is ³S₀ and is nonmagnetic. A simplified Grotrian diagram of Hg is shown in Fig. 3. Excitation to the ${}^{3}P_{1}$ state is achieved by illumination with a light source near 254 nm, and the next step is to populate the long-lived metastable state ${}^{3}P_{2}$. The simplest way to accomplish the optical pumping is with a mercury UV lamp, filtering the green line at 546 nm, which will then cause atoms to accumulate in the upper metastable state. This filtering can be done with a band-reject coating on a UV-grade fused-silica vacuum tube. An array of UV lamps would be prepared with a pure isotope of Hg, so that the emitted lines would only excite those atoms inside the vacuum chamber. An alternative that will be explored is to load the lamps with the desired isotopic mixture so that the excited Hg atoms will already reflect this mixture. Such a method can work in the case of Hg because the isotope shifts are considerably larger than the Doppler shifts.

C. Magnetic Guiding and Collection

After optical pumping, the atoms enter a magnetic separation region. Since the essence of our magnetic separation scheme can

be 2-D as explained in the following, it means we are in fact not restricted to 3-D cylindrical multipole guides proposed earlier [16]. We may, therefore, consider a whole set of magnet geometries that are made of planar arrays that only act in the plane of the page but leave the atoms to travel ballistically into and out of the page. Such magnetic deflection and reflection geometries can in many cases make the apparatus simpler and more efficient. One example of such magnetic geometries would use planar Halbach arrays [17] rather than cylindrical multipole arrays. Halbach arrays maximize the field on one side of the array and minimize it on the other, with the range of the field being on the order of the size of the magnets. The force F applied to the atoms by the magnets depends on the value of the atoms magnetic quantum number and the gradient of the magnetic field magnitude B (primarily toward the magnet surface) by $F = -\mu_B g_I m_J B$, where $\mu_{\rm B}$ is the Bohr magneton, and g_J is the Landé g-factor. This typically leads to accelerations around 10–100 km·s⁻² for gradients of 1 T \cdot cm⁻¹ achievable with available rare-earth permanent magnets. Here, we will consider three such planar guide configurations depicted in Fig. 4. The first configuration (C1) consists of two Halbach arrays facing each other, forming a trough with magnetic barriers at each wall. This forms a guide whose cross section in the plane of the paper would look similar to the cylindrical guide cross section (though with some field modulation in the direction of the beam). This is useful in the case of multistage guiding, since the atoms would exit the first guide with well-defined positions and trajectories in the guiding plane. The second configuration (C2) would be that shown in Fig. 4. The arrays create a series of simple curved barriers always in the direction of the curve. Since line of sight is blocked, and there is very little field on the back side of the arrays, only low-field-seeking atoms can be deflected away from the magnets and through the guide. The disadvantage of these first two methods is that on average, only 40% of the atoms will decay into a magnetic state that can be guided, m = 1 or m = 2states in the ³P₂ manifold. The third configuration (C3) would be similar to C1, in that two arrays would face each other, but the planes would not be curved. Instead, there would be a thin wall at the field minimum in the center and at the maximum by the magnets, and there is no line of sight to the walls from the source. Therefore, any nonmagnetic atoms (or atoms going too fast) would fly straight through. Atoms with a negative mj would collect on the magnet walls and those with a positive mj would collect on the center wall. This configuration is ideal for mercury with the above optical pumping by a lamp, since it could capture about 80% of the desired isotopes, all those except for the m=0 state in the 3P_2 manifold.

IV. SUMMARY

We estimate that the cost of Hg isotope separation could be less than \$100 per gram of an optimized mixture, which implies an added cost of 30 cents for a linear 4-ft fluorescent lamp at the current dose per lamp. The energy savings over the lifetime of the lamp would be around \$30, so there is a compelling business model. The next step is to test lamps with the optimized mixtures in order to validate the Monte Carlo simulations. Following a successful test, a pilot separation machine will need to be built, demonstrating cost-efficient separation of Hg isotopes using MAGIS.

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