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Magneto-optical isotope enrichment of Potassium-40 with transverse cooling

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Isotope shifts among different isotopes can be effectively addressed using narrow-linewidth lasers, facilitating laser isotope separation and achieving significant enrichment at a single stage. The separation of potassium isotopes, employing optical pumping and magnetic deflection, has proven to be efficient. To further improve the enrichment of ⁴⁰K, we introduce 2D transverse cooling to minimize the divergence angle. Through this modification, we demonstrate enrichment of ⁴⁰K, elevating it from 0.012% to 12% - 20%. This represents an enrichment increase by three orders of magnitude, surpassing our previous result by one order. Our method is particularly well-suited for isotope enrichment of elements with extremely low abundance.

Keywords: isotope separation, potassium isotopes, 2D transverse cooling. **DOI:** xxxxxx/COLxxxxxx.

1. Introduction

Rare isotopes are very important and useful in many aspects, such as date tracing^[1,2], precision measurement^[3,4], and quantum simulations^[5,6]. In cold atom physics, ⁴⁰K is an important isotope that is widely used to study the quantum degenerate Fermi gas^[7]. But the natural abundance of ⁴⁰K is extremely low, about 0.012%, which is hard to use in the lab. The widely used K source is extracted from nuclear waste with an abundance of about a few percent for ⁴⁰K, which is far away from the natural abundance, and the typical weight is about a few milligrams. It is expensive and the supply is not stable, a convenient separation method to achieve such a goal will be very useful. Traditional separation methods, such as the centrifuge and chemical method, are unsuitable for this extremely low abundance isotope. Laser isotope separation turns out to be a feasible method, which can be used to produce small mount samples with a high enrich factor such as 100 -1000.

Laser isotope separation makes use of the isotope shift, which has typical values of a few hundred MHz to GHz. Although it is small, it can be easily addressed by those narrow linewidth lasers^[8–12]. This idea was extensively explored in the 1980s but only succeeded with small mount samples. In the last decade, an optical magnetic combined method was developed for isotope separation and enrichment^[13]: atoms with different isotopes are first optically pumped to different spin states, then enter a magnetic field regime with a high gradient. Atoms in low-field seeking states can be deflected and then collected, thus achieving isotope separation. The optical pumping efficiency is so high, resulting in a high purity of isotopes. Those methods have been demonstrated in both $Li^{[14,15]}$ and $K^{[16]}$.

In our prior research, we effectively utilized the magnetooptical method to separate potassium isotopes, achieving a production rate of 1.1 µg/day and enriching ⁴⁰K to approximately 1% abundance^[16]. While this already presents a significant enhancement of nearly 100 times the natural abundance, it is still ten times lower than our desired goal. A critical limitation in our previous methodology is the presence of a divergence angle. Despite employing a micro-capillary array in the oven to pre-collimate the atomic beam, a residual divergence angle of about 3 degrees persisted^[17–19]. Because a long magnetic wall (about 2 meters) is used to separate the low-field seeking and high-field seeking states, even a small divergence angle still results in a large spreading of atoms and thus loss. Although implementing a collimation hole could potentially reduce the divergence angle, it would also curtail the production rate.

The advancements in laser cooling techniques offer a promising solution by enabling the reduction of divergence angles through two-dimensional (2D) transverse cooling. Laser cooling easily achieves a transverse temperature lower than millikelvin^[20-24], corresponding to a divergence angle of a few milliradians. This enhancement can lead to a one-order-ofmagnitude improvement in separation efficiency.

It is noteworthy that while 2D transverse cooling has been widely used in cold atom physics, its application in laser isotope separation remains unexplored. An essential consideration is its impact on production rates. In our scenario, the pivotal



Fig. 1. (a) The diagrammatic sketch for the Potassium isotope separation machine. Atoms are emitted from a heated oven, collimated by the light in the 2D region, and pumped by L_1 and L_2 . A curved magnet array deflects atoms with the right magnetic moment. And finally, atoms are imaged by the probe laser L_p . (b) Enlarged view of the 2D region where the particles are collimated. (c) The ⁴⁰K fluorescence we took in the detection area. The top and bottom images were taken by the camera, and the y-axis and z-axis represent the position of the pixels in the photo. The top one has no light in the 2D region, and the bottom one has 2D transverse cooling.

step is optical pumping, known for its high efficiency, requiring only a minimal number of photons to induce spin flips and separation^[13,15,16]. Conversely, 2D transverse cooling necessitates several hundred to a thousand photons for beam collimation, which may reduce the production rate significantly. However, in our specific context, the natural abundance of ⁴⁰K is approximately 600 times lower than that of ⁴¹K, and 8000 times lower than ³⁹K. Therefore, it's the photon count associated with ³⁹K that limits the production rate, not ⁴⁰K, even with 2D transverse cooling. Our approach holds particular promise for rare isotopes with exceptionally low abundance.

2. Experiment setup

Figure 1 (a) illustrates the experimental setup. The atomic beams are collimated using a micro-capillary array, consisting of 400 needles with an inner diameter of 0.3 mm and a length of 10 mm^[17]. Post oven, atoms undergo 2D transverse cooling (along *x* and *y*) facilitated by a laser operating at the *D*2 transition at 767 nm. This laser comprises two frequencies: a transition from F = 9/2 to F' = 11/2 and another from F = 7/2 to F' = 9/2. These frequencies are well-separated from the transitions of ³⁹K and ⁴¹K, with a minimum detuning of 369 MHz and 264 MHz, respectively. Importantly, our experimental observations indicate that the addition of 2D lasers has minimal impact on the signal of ³⁹K and ⁴¹K.

Subsequently, the atoms are optically pumped to high-field seeking states for ³⁹K and ⁴¹K, and low-field seeking states for ⁴⁰K using narrow linewidth lasers (L_1 and L_2). These lasers

are stabilized by DAVLL (Dichroic-atomic-vapor laser lock), with a line width of 500kHz. The polarized potassium atoms then enter the 1.5-meter-long Halbach array with a maximum magnetic gradient of 2.5 T/cm near the magnet surface. In this configuration, only low-field seeking atoms are deflected, enabling the deflection and collection of ⁴⁰K. For more details please refer to our previous work^[16].

Figure 1 (b) shows an enlarged view of the 2D region where the particles are collimated. In the transverse cooling regime, the laser beam has an elliptical size of 15 mm \times 7.5 mm, undergoing more than ten reflections by mirrors with a total transmission rate of 0.9 in each cycle, resulting in an effective cooling regime of 14.5 cm. The two mirrors are positioned at an angle of approximately 0.05 degrees. Initially, the incidence beam has an angle of about 2 degrees, gradually becoming nearly perpendicular to the mirrors. Such a small angle allows light to bounce back and forth between the two flat mirrors and spread the light spot as far as possible over the entire 2D area. On the other hand, because the incident angle of 2D light is gradually decreasing, it can match the velocity direction of atoms over a long distance and also match the velocity direction of different atoms. This configuration was widely used in the early study of laser cooling of atoms^[24-26] and more recently of laser cooling of molecules.

Laser cooling typically reduces the transverse temperature to a few microkelvins, implying a divergence angle of just a few milliradians. Since the 2D transverse cooling is exclusively applied for ⁴⁰K, resulting in highly collimated ⁴⁰K atoms, while ³⁹K and ⁴¹K atoms remain unaffected, this leads to a further enhancement in isotope enrichment. Figure 1 (c) displays typical data of ⁴⁰K fluorescence captured in the detection area. The upper panel depicts data without 2D transverse cooling, whereas the lower panel incorporates 2D transverse cooling. An enhancement of approximately 20 times is observed with the application of 2D transverse cooling.

3. Numerical simulation

To study the enhancement effect with 2D transverse cooling, we applied a numerical simulation based on the multi-level rate equations^[27,28], and we briefly summarize here. The populated fraction in each sublevel for a multi-level system is calculated as

$$\frac{dN_l}{dt} = \Gamma \sum_{u} r_{l,u} N_u + \sum_{u,p} R_{l,u,p} (N_u - N_l),$$

$$\frac{dN_u}{dt} = -\Gamma N_u + \sum_{l,p} R_{l,u,p} (N_l - N_u),$$
(1)

where N_l and N_u are the populated fractions for the *l*-th sublevel in the ground state and the *u*-th sublevel in the excited state, Γ is the spontaneous decay rate of the excited state, $r_{l,u}$ is CG factor.

$$R_{l,u,p} = \frac{\Gamma}{2} \frac{r_{l,u} s_p}{1 + (2\Delta_p / \Gamma)^2}$$
(2)

is the excitation rate for $l \rightarrow u$ transition from the *p*-th laser beam, and s_p is the saturation factor and Δ_p is the detuning.



Fig. 2. (a) The trajectories of particles after exiting the oven. θ represents the divergence angle at which the particle velocity deviates from the main axial direction. (b) shows the numerical results of ⁴⁰K enhancement versus the laser frequency. The total intensity of 2D laser is 280 mW/cm², and the oven temperature is 600 K. Square markers result from numerical simulation, and the red line is the Gaussian fitting curve. (c)shows the numerical results of ⁴⁰K enhancement versus the total intensity of 2D laser. The laser detuning is 5MHz and the oven temperature is 600 K. Square markers result from numerical simulation, and the red line is the fitting curve with the function $y = A \times I/(I + I_s)$. (d) The divergence angle of the particle beam leaves the 2D region with (red) and without (blue) transverse cooling. The laser detuning is 5 MHz, the total intensity of the 2D laser is 280 mW/cm² and the oven temperature is 600 K. The green dotted line represents 0.38 degrees, with only particles having a divergence angle less than this value likely to enter the detection region.

Numerically, we simulate numerous potassium atoms with random positions, random velocity, and two random angles, allowing us to determine velocities in all three directions. The initial position is confined within a size of 6 mm× 6 mm set by the furnace. One angle spans from 0 to 2π , while the other is more restricted, as discussed below. The cooling force is computed following our previously described methodology. Atomic trajectories are then calculated accordingly with the geometry constraint of our experiment setup.

Figure 2(a) displays typical trajectories of particles after their exit from the source. We define the divergence angle as the deviation angle of particle velocity from the main axial direction. Due to the mechanical limitations of the vacuum chamber, the actual divergence angle is capped at 0.38 degrees. Only atoms with divergence angles smaller than this value are eventually counted. And finally, we analyze the distribution and count the number of atoms at the detection regime. By calculating the ratio of the number of 40 K atoms with and without 2D transverse cooling, we derive the enhancement of 40 K.

Figure 2(b) and (c) displays the dependence of 40 K enhancement on the laser detuning and intensity of the 2D laser. Maximum enhancement is observed in Fig. 2(b) when the detuning is around 5 MHz with the total intensity 280 mW/cm² and the oven temperature 600 K. Despite the usual requirement

for red-detuned lasers in laser cooling, we need to use a bluedetuned laser in our experiment due to the small angle between our laser and the atomic beam as shown in Fig. 1(b). According to the fitting function $y = A \times I/(I+I_s)$, we get that the enhancement of potassium reaches saturation at around 100 mW/cm², when the detuning is 5 MHz and the oven temperature is 600 K, as shown in Fig. 2(c). Notably, the saturation intensity of potassium is $I_{sat} = 1.75$ mW/cm², and the simulated saturation intensity is 30 times that value. We suspect that the reason for this discrepancy may be due to the initial collimation of particles by laser. A relatively high intensity helps to collimate particles faster, and more likely they will enter the detection region.

Figure 2(d) shows the divergence angle of the atomic beam leaving the 2D region with and without 2D transverse cooling. When 2D transverse cooling is switched on (red line), the divergence angle is reduced significantly, which increases the 40 K atoms by one order of magnitude.

4. Data and analysis



Fig. 3. (a) The ⁴⁰K signal versus the laser frequency at different temperatures ranging from 220 °C to 300 °C with the total intensity 260 mW/cm². The curve represents the Gaussian fitting. (b) The ⁴⁰K signal versus the laser intensity with detuning 5 MHz. The curve is the fitting curve with the function $y = A \times I/(I + I_s)$.

Figure 3 illustrates the flux rate of 40 K at the detection area at varying temperatures, with the total intensity of 260 mW/cm². A notable enhancement occurs when the detuning is within

5–8 MHz, as shown in Fig. 3(a). The slight offset detuning is attributed to the small angle between the laser and the atomic beam^[25]. Center frequencies exhibit a slight shift with increasing the oven temperature, influenced by the varying forward velocities connected with the temperatures. Figure 3(b) displays the intensity dependence of ⁴⁰K signal, which starts to saturate around 150 mW/cm², reaching a flux rate of $2.8 \times 10^{10}/s$ at 300 °C.



Fig. 4. (a)The enriched abundance of 40 K at different temperatures (black triangle) and the gain from 2D transverse cooling (red square). (b) The suppression ratios for both 39 K and 41 K.

Figure 4(a) explores the temperature dependence of the enriched abundance of 40K and the gain from 2D transverse cooling under saturated intensity and optimal detuning. Abundance was calculated by measuring the signal intensity of ⁴⁰K divided by the signal intensity of ⁴⁰K plus the signal intensity of ³⁹K and the signal intensity of ⁴¹K when all light was turned on. With the increase of temperature, the abundance reaches more than 20% at 240 °C, and then gradually decreases to 12% at 300 °C. Gain, defined as the signal ratio with and without the 2D transverse cooling, increases ⁴⁰K production rate by 17 to 22 times across the temperature range from 220 °C to 300 °C. This aligns closely, albeit slightly higher, with theoretic prediction, reinforcing the significance of the divergence angle in the final production rate. At higher temperatures, the gain exhibits a slight reduction. Two potential reasons are considered: first, the optimized conditions for 2D transverse cooling at lower temperatures may not be ideal at higher temperatures due to increased forward atom velocities; second, higher temperatures result in elevated K atom density, leading to increased scattering and a potential decrease in optical pumping efficiency. Figure 4(b) shows the suppression ratio for both ³⁹K and ⁴¹K. The suppression ratio, indicating the atomic output with and without optical pumping, remains nearly constant for ⁴¹K, while for ³⁹K, it decreases with rising temperature. The decrease is attributed to reduced optical pumping efficiency due to scattering light from 2D transverse cooling and optical pumping regimes. Given the high natural abundance of ³⁹K, the decrease in the suppression ratio correlates with a decline in the abundance of enriched ⁴⁰K, as shown in Fig. 4(a).

5. Discussions and Conclusion

Collimation of the atomic beam is crucial for our laser isotope separation method, and we have found that 2D transverse cooling effectively addresses this issue. The magnetic isotope separate stage has some phase space (position-velocity) acceptance region. Typically, we regulate the total velocity by adjusting the temperature. However, the average speed \overline{v} proportional to $T^{1/2}$ changes minimally in the experiment's temperature range (220 °C-300 °C), varying from 508 m/s to 548 m/s. Conversely, the reduction in transverse velocity (divergence angle) with 2D transverse cooling allows more atoms to enter the acceptance region that would otherwise be excluded. Our final experimental results confirm that reducing the divergence angle leads to one order of magnitude improvement. Specifically, at a temperature of 300 °C, we achieved a potassium production rate of 1.3 μ g/day, with approximately 12% of it being ⁴⁰K. This abundance is particularly beneficial for cold atom experiments involving 40K. Moving forward, our future plans involve optimizing the needle array and furnace design to further increase the atomic flow rate and production rate for potassium isotopes.

One of the primary limitations we face is the photon number scattered in relation to ³⁹K, not ⁴⁰K, due to the extremely low natural abundance of ⁴⁰K. The average forward speed \bar{v} is approximately 550 m/s when the oven is 300 °C. Considering the divergence angle θ of the atomic beam is 2 degrees, the transverse velocity is

$$v_{\perp} = \overline{v} \times \tan \theta = 19.2 \, m/s \tag{3}$$

The recoil velocity v_{rec} of 40 K is 1.3 cm/s. Therefore, the approximate number of photons that need to be scattered in 2D transverse cooling is

$$N = v_{\perp} / v_{\rm rec} \approx 1500 \tag{4}$$

The natural abundance of 39 K is 8000 times higher than 40 K, given the same intensity of lasers for both 39 K and 40 K, the final production rate of enriched potassium is still restricted by the photon number scattered by 39 K.

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