

Proposal for the generation of continuous-wave vacuum-ultraviolet laser light for Th-229 isomer precision spectroscopy

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Laser spectroscopy of the ^{229}Th isomeric transition has recently emerged as a field with several groundbreaking advancements. However, the employed vacuum-ultraviolet (VUV) laser sources in these studies are all pulsed, resulting in low spectral power densities that are largely inadequate for coherently driving this extraordinarily weak isomeric transition. Here, we propose generating a cw laser at 148.4 nm using four-wave mixing (FWM) in cadmium vapor. The large transition-matrix elements of cadmium, based on state-of-the-art electron-correlation calculations, and the optimized FWM resonance-enhanced scheme with readily accessible incident laser wavelengths, lead to a giant third-order nonlinear susceptibility. Combined with the high coherence of the FWM process, cadmium vapor emerges as a promising nonlinear medium capable of producing a cw VUV laser with over 30- μW power and a narrow linewidth. This approach addresses the lack of an intense and narrow-linewidth laser near the ^{229}Th isomeric transition, allowing for the coherent driving of nuclear Rabi oscillations and advancing the development of the nuclear optical clock.

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I. INTRODUCTION

The invention of coherent laser sources has revolutionized the field of atomic and molecular physics. The success of these applications can be attributed to the ability to drive transitions in the electron shell using lasers. Expanding these well-established technologies to manipulate nuclear quantum states holds great promise. However, coherent control of nuclear transitions has been challenging [1–3] due to the significant energy gap between the nuclear transitions and the photon energy of coherent laser radiation. A nuclear transition involves large changes of both Coulomb interaction energy and strong interaction energy that are normally a few 10^5 – 10^6 electron volts. A fortuitous cancellation of the changes of these two terms occurs in the well-known ^{229}Th isomeric transition [4,5], resulting in a nuclear excited state with the lowest known energy that lies only 8.4 eV above the ground state [6–15]. The

corresponding transition falls within the range of typical electronic transitions, and is the only nuclear transition that can be driven using currently available laser sources. This offers a unique opportunity to bridge the fields of atomic and nuclear physics and potentially apply quantum technologies developed over the past few decades to nuclei.

One remarkable frontier is the development of a nuclear optical clock [16,17]. Due to their much smaller size and multipole moments compared to atoms, nuclei exhibit greater robustness in the frequency of nuclear transitions against perturbing external fields. This characteristic can be employed to alleviate the significant effort required to control ambient fields near an atomic optical clock. The use of the ^{229}Th isomeric transition in a nuclear clock is further motivated by its remarkable sensitivity to temporal variations in fundamental constants, such as the fine-structure constant [4,18–21], and its application in the search for ultralight dark matter [22,23].

Another salient feature of nucleus-based experiments is the doping of a large number of atoms into a solid material while generically keeping the nuclear transitions intact

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[16,24,25]. This is in contrast to current atomic-shell-based quantum systems, where valence electrons of suspended atoms in the vacuum play a central role, and doping would eliminate the relevant underlying quantum states. Hosting nuclei within a solid material in the Lamb-Dicke regime may eliminate the need for laser cooling and trapping, thereby simplifying the experimental efforts. Additionally, the ability to control a macroscopic quantity of nuclear systems can largely reduce the interrogation time for precision measurements.

Driven by these compelling prospects [26–28], several methods have been employed to measure the energy of the ^{229}Th isomeric transition [6–15], including indirect γ spectroscopy [8,11,12], internal-conversion electron spectroscopy [9,29], and vacuum-ultraviolet (VUV) spectroscopy of radiative fluorescence light [13]. Very recently, laser spectroscopy has been finally achieved by two groups based on thorium-doped VUV-transparent crystals [14,15], and the transition wavelength is determined to be 148.4 nm. In both experiments, tunable-pulsed VUV laser sources are produced by four-wave mixing (FWM) in xenon gas, with durations of several nanoseconds, average powers ranging from 30 to 450 μW , and linewidths of a few GHz. The spectroscopy uncertainty of these two studies stands at approximately 3 GHz [14,15], primarily limited by the broad spectral linewidth associated with the pulsed nature of the VUV laser. The radiative lifetime of the isomer state in vacuum is measured to be between 2000 and 3000 s [13–15,21,30], corresponding to a linewidth of around 60 μHz .

This significant linewidth disparity, ranging from GHz in pulsed VUV laser to μHz in the transition linewidth, underscores the necessity for a narrow-line laser for efficient transition driving and precision spectroscopy. A highly coherent frequency comb in the VUV region, generated via high-order harmonic generation (HHG) from an infrared comb, has been demonstrated [31–33] and utilized in ^{229}Th isomer spectroscopy [27,34–36]. Although the comb linewidth might be as narrow as kHz, only a single comb line, with a power of 1 nW, contributes to driving the isomeric transition [27,36,37]. Phase-matched frequency conversion in nonlinear crystal near 150 nm has also been achieved [38]. However, due to strong absorption and dispersion, the conversion efficiency rapidly drops to near zero below 150 nm, even for pulsed lasers with extremely high peak intensity [38,39].

In this work, we propose generating cw laser light at 148.4 nm using four-wave mixing in cadmium vapor for precision spectroscopy of the ^{229}Th isomeric transition. Cadmium is chosen for its large third-order susceptibility and the availability of high-power fundamental lasers near 375.2 and 710.1 nm. Together with its relatively low melting point, which supports high vapor pressure at reasonable temperatures, cadmium vapor provides a practical and efficient medium, capable of generating over 30 μW

of cw VUV radiation at 148.4 nm. This output is more than 4 orders of magnitude higher than a single comb line in state-of-the-art VUV frequency combs [36,37]. We argue that FWM is highly phase coherent and has the potential to support a VUV laser linewidth several orders of magnitude narrower than the pulsed FWM sources currently used in isomer spectroscopy [40,41]. This development paves the way for coherently driving the ^{229}Th isomeric transition and the construction of the nuclear optical clock [42].

II. THEORY OF THE FWM PROCESS IN CADMIUM VAPOR

The FWM process relies on the third-order nonlinear susceptibility, which is the lowest-order nonlinearity observed in an isotropic medium. It has long been employed to generate VUV light [43,44] that otherwise is challenging to produce for spectroscopy in this wavelength region. The nonlinear process necessitates high incident laser intensities, generally resulting in the generated VUV light being pulsed. A notable exception is the production of cw Lyman- α radiation at 121.6 nm using FWM in mercury vapor [45–48].

For the FWM process in cadmium vapor considered here, another wave with the sum frequency of three incident laser beams is generated, $\omega_1 + \omega_2 + \omega_3 \rightarrow \omega_4$, as shown in Fig. 1 for the special case where $\omega_1 = \omega_2$. For tightly focused incident laser beams with equal confocal parameter b , the FWM output power in the small signal

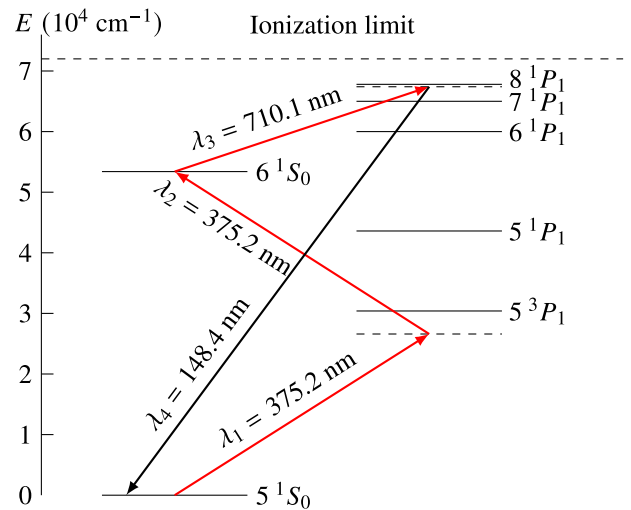


FIG. 1. Simplified energy levels of neutral cadmium in terms of wave number. The arrows represent incident lasers with wavelengths λ_1 , λ_2 , λ_3 and the generated VUV laser with wavelength λ_4 at 148.4 nm.

limit [44] is given by [47,49,50]

$$P_4 = \frac{9}{4} \frac{\omega_1 \omega_2 \omega_3 \omega_4}{\pi^2 \epsilon_0^2 c^6} \frac{1}{b^2} \left(\frac{1}{\Delta k_a} \right)^2 |\chi_a^{(3)}|^2 P_1 P_2 P_3 G(bN \Delta k_a), \quad (1)$$

where ω_i is the laser angular frequency, P_i the laser power, N the number density of the nonlinear medium, $\Delta k_a = (k_4 - k_1 - k_2 - k_3)/N$ the wave vector mismatch per atom, $\chi_a^{(3)}$ the third-order nonlinear susceptibility per atom, and $G(bN \Delta k_a)$ the phase-matching function.

To understand the FWM process described by Eq. (1), the phase-matching function $G(bN \Delta k_a)$ and the calculation of nonlinear susceptibility $\chi_a^{(3)}$ are detailed in the following.

The wave-vector mismatch Δk_a due to dispersion indicates the momentum mismatch in nonlinear optical processes. In the tight focus limit, where the confocal length of the laser beams b is much smaller than the length of the nonlinear medium L , this mismatch can be compensated by the unbalanced Gouy phase shift. In this limit, the Gouy phase shift is experienced three times by the incident beams in the Gaussian focus but only once by the frequency-summed generated beam [47]. Thus, the phase-matching function $G(bN \Delta k_a)$ is maximized not at $\Delta k_a = 0$ as in the nonlinear optical processes with the collimated beams, but at $bN \Delta k_a = -4$ [49]. It is worthwhile to note that Δk_a does not depend on the number density of the nonlinear medium (see Appendix B for more details), but is fully determined by the frequencies of the incident and generated lasers, as well as the transition strengths of the cadmium atom. Therefore, the optimal phase-matching condition can be achieved by adjusting b and N accordingly [49].

The calculation of the nonlinear susceptibility $\chi_a^{(3)}$ from the first principles can be found in Ref. [51]. We employ the two-photon resonance condition, $\omega_1 + \omega_2 = \omega_{rg}$, to enhance the output VUV power [43], with 6^1S_0 as the resonance state (denoted as r , with ω_{rg} representing its transition frequency to the ground state g). For three linearly polarized incident laser beams, the leading term in $\chi_a^{(3)}$ is [50]

$$\chi_a^{(3)} = \frac{1}{6\epsilon_0 \hbar^3} S(\omega_1 + \omega_2) \chi_{12} \chi_{34}, \quad (2)$$

with

$$S(\omega_1 + \omega_2) = \frac{1}{\Omega_{rg} - (\omega_1 + \omega_2)}, \quad (3)$$

$$\chi_{12} = \sum_l \left(\frac{\mu_{rl} \mu_{lg}}{\omega_{lg} - \omega_1} + \frac{\mu_{rl} \mu_{lg}}{\omega_{lg} - \omega_2} \right), \quad (4)$$

$$\chi_{34} = \sum_m \left(\frac{\mu_{rm} \mu_{mg}}{\omega_{mg} - \omega_4} + \frac{\mu_{rm} \mu_{mg}}{\omega_{mg} + \omega_3} \right). \quad (5)$$

Here, $\Omega_{rg} = \omega_{rg} - i\Gamma_r/2$ is the complex transition frequency, containing the transition frequency ω_{rg} and the total population decay rate of the r state Γ_r , and μ_{ij} are the z components of the transition-matrix elements coupling states i and j . The summation is performed over all the intermediate states $l(m)$ (namely, states $n^1,3P_1$), whose transition frequencies to the ground state g are denoted as $\omega_{lg}(\omega_{mg})$.

$S(\omega_1 + \omega_2)$ describes the shape of the two-photon resonance. In Eq. (3), pressure broadening and Doppler broadening is not considered for simplicity, although both effects are included in the final analysis. The pressure broadening $\Delta\omega_p$ can be included in the expression of the imaginary part of Ω_{rg} by $\Gamma'_r = \Gamma_r + 2\Delta\omega_p$ [51]. The Doppler broadening, which contributes through the Doppler shift of ω_1 and ω_2 , can be introduced by a convolution of $S(\omega_1 + \omega_2)$ with the Maxwell-Boltzmann velocity distribution function (see Appendix C for a full mathematical treatment), and dominates the linewidth of $S(\omega_1 + \omega_2)$. Both broadening effects reduce $S(\omega_1 + \omega_2)$ and, consequently, the FWM conversion efficiency.

III. RESULTS

A. Nonlinear susceptibility and output power

For experimental simplicity, we assume $\omega_1 = \omega_2$, where only two incident laser beams at 375.2 and 710.1 nm are required to produce the VUV light (see Fig. 1). For the more general case where $\omega_1 \neq \omega_2$, see Appendix F.

For cadmium vapor with natural abundance, $S(\omega_1 + \omega_2)$ should be calculated for each isotope and weighted according to their relative abundances in the summation [47,50]. To our knowledge, the isotope shifts (IS) of the $5^1S_0 - 6^1S_0$ transition in cadmium have not been measured. We calculate the corresponding field- and mass-shift factors for the $5^1S_0 - 6^1S_0$ transition using the configuration interaction method with many-body perturbation theory (CI+MBPT) with inclusion of single- and double-core excitations (see Table I in Appendix C). Along with the difference in the mean-square charge radius extracted from the available experimental data on the $5^1S_0 - 5^3P_1$ transition [52], we find that IS of the $5^1S_0 - 6^1S_0$ transition are much smaller than the linewidth of $S(\omega_1 + \omega_2)$, and using pure isotope only marginally increases the output VUV power (see Appendix C). We assume the use of pure ^{114}Cd isotope for simplicity in further computations.

Theoretically, the summation in Eqs. (4) and (5) is carried out over an infinite number of intermediate states. In our calculation, however, the summation is done over 20 intermediate P states, namely n^1P_1 and n^3P_1 states with $n = 5 - 14$, since the transition-matrix elements for high-lying states decrease substantially and the transition frequencies of the corresponding states are detuned from the generated VUV light. Considering the dipole

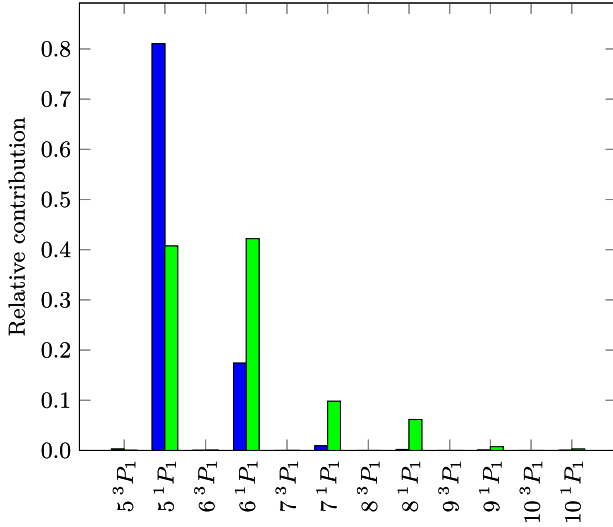


FIG. 2. The normalized relative contributions to χ_{12} (blue bars) and χ_{34} (green bars) from a set of intermediate P states up to the 10^1P_1 .

approximation and assuming linear laser polarization, the selection rules for the coupled states are $\Delta J = \pm 1$ and $\Delta M_J = 0$. All corresponding matrix elements are calculated (see Table II in Appendix) using the relativistic Fock-space coupled cluster (FS-CC) method implemented in DIRAC [53,54] and Exp-T [55,56] program packages. Our results are consistent with theoretical and experimental data for available transitions involving low-lying excited states ($5^1S_0 \rightarrow 5^3P_1$, $5^1S_0 \rightarrow 5^1P_1$, $5^1S_0 \rightarrow 6^1P_1$, $5^1P_1 \rightarrow 6^1S_0$, and $5^1P_1 \rightarrow 5^1D_2$), with a discrepancy of no more than 10% [57–60]. A more detailed investigation of relativistic and electron-correlation effects for high-lying states is presented in Ref. [61].

The relative contributions to χ_{12} and χ_{34} from various intermediate P states are shown in Fig. 2. The dominant contributions to χ_{12} and χ_{34} originate from the 5^1P_1 and 6^1P_1 states. For χ_{34} , the 7^1P_1 and 8^1P_1 states each contribute approximately 10%. Contributions from all triplet states and states above the 8^1P_1 to both χ_{12} and χ_{34} are minor.

To explore the tunability of our source, we choose incident laser powers of 3 W at 375.2 nm and 6 W at the third photon wavelength λ_3 , which varies from 648.6 to 844.8 nm. These powers are readily available in the laboratory. The confocal parameter is set to $b = 1.5$ mm for experimental convenience, and, accordingly, the vapor temperature is set to $T = 580^\circ\text{C}$ to optimize the output VUV power. The saturation vapor pressure as a function of temperature can be found in Fig. 12 in the Appendix. Under these conditions, we calculate the nonlinear susceptibility $\chi_a^{(3)}$, the phase-matching function $G(bN\Delta k_a)$, and the generated VUV power P_4 in the vicinity of ^{229}Th isomeric transition, as shown in Fig. 3. The nonlinear susceptibility $\chi_a^{(3)}$ exhibits divergence near resonant states

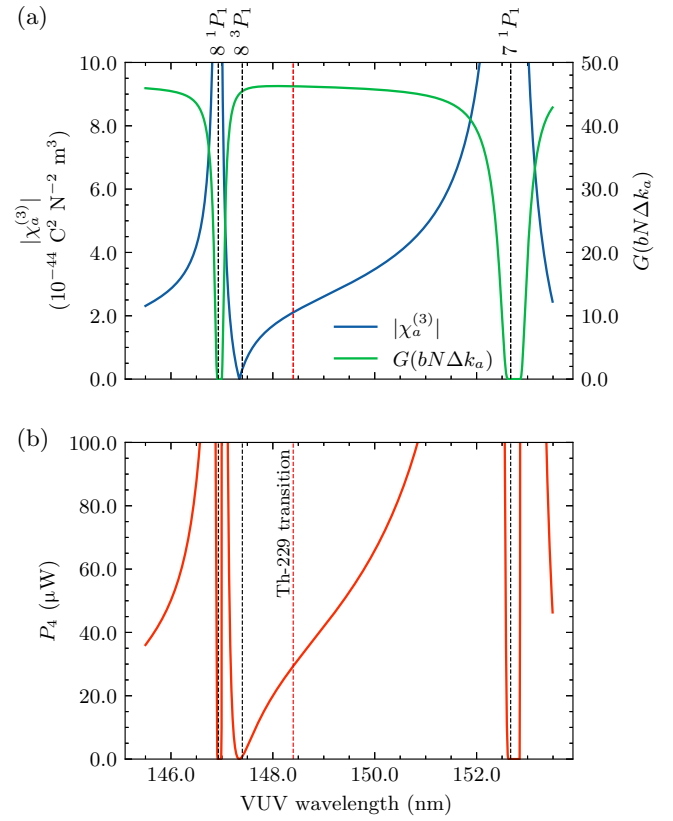


FIG. 3. (a) Nonlinear susceptibility $|\chi_a^{(3)}|$, phase-matching function $G(bN\Delta k_a)$, and (b) the VUV output power P_4 in the vicinity of ^{229}Th isomeric transition according to Eq. (1) using parameters stated in the text. The VUV wavelength range (145.5–153.5 nm) on the horizontal axis corresponds to varying λ_3 from 648.6 to 844.8 nm. Note that the effects of absorption and saturation are ignored in the plot.

since dissipative terms are not accounted for in Eqs. (4) and (5). The phase-matching function $G(bN\Delta k_a)$ goes to zero near resonant states because the refractive index of the generated beam is so large that the resulting wave-vector mismatch Δk_a cannot be compensated for using the current parameters (see Appendix for how phase matching can be restored). Zero VUV power points arise (e.g., near 147.3 nm) when terms with different signs cancel in the expression for χ_{34} given by Eq. (5). The output power P_4 at 148.4 nm is approximately $30 \mu\text{W}$.

Conservatively, the VUV light can be generated within the 140–176 nm range, corresponding to third-photon wavelengths from $\lambda_3 = 552$ nm to $\lambda_3 = 2851$ nm (see Appendix G). This range may be further extended below 140 nm by leveraging the ionization continuum and autoionizing resonances [62] of cadmium, and beyond 176 nm by employing third-photon wavelengths exceeding 2851 nm.

We also perform calculations for the FWM scheme exploiting 5^1D_2 as the two-photon resonance state, which

is described in detail in Appendix A. Similar to the case with 6^1S_0 as the resonance state, the coupling to 5^1P_1 and 6^1P_1 states dominates the nonlinear susceptibility, and the output VUV power is at a comparable level.

B. Output power enhancement and saturation

The VUV power can be further enhanced by introducing a noble gas to provide additional compensation for wave-vector mismatch, or by tuning one of the incident beams close to the $5^{1,3}P_1$ resonance states, while maintaining two-photon resonance with the second incident beam. For a detailed discussion, see Appendixes E and F.

The output VUV power in Fig. 3(b) does not account for absorption and saturation effects. Saturation might be relevant in the tightly focused case, where the laser intensity near the beam waist can be enormous. We find that the only significant saturation effect under the current experimental parameters is due to the depletion of the ground state induced by the two-photon resonance Raman-type absorption. This issue can be mitigated either by slightly detuning the lasers from the exact two-photon resonance condition, or by loosening the focus while adding some noble gas into the medium for phase-mismatch compensation. Other effects, such as the linear absorption of the beams, other third-order and higher-order nonlinear processes, and amplified spontaneous emission are found to be insignificant. For more details, see Appendix D.

IV. DISCUSSION

A. Nuclear Rabi oscillation condition

As a conservative estimate, we assume a VUV power of $10\ \mu\text{W}$ focused to a spot diameter of $1.8\ \mu\text{m}$ to study the interaction of the ^{229}Th nucleus with the VUV light. This interaction has been treated in detail in Ref. [63]. We consider the ^{229}Th isomeric transition as the simplest two-level system interacting with a VUV laser with intensity I_l and linewidth $\Gamma_l/2\pi$. In order to observe nuclear Rabi oscillations, the linewidth of the VUV laser must satisfy the condition [63]

$$\Gamma_l < 2\Omega_{eg} = 2\sqrt{\frac{2\pi c^2 I_l \Gamma_\gamma}{\hbar \omega_0^3}} \approx 2\pi \times 1\ \text{kHz}, \quad (6)$$

where Ω_{eg} is the Rabi frequency, Γ_γ the radiative decay rate of the nuclear excited state, and ω_0 the angular frequency of the isomeric transition. The nuclear excitation probability as a function of VUV irradiation time for laser linewidths below and above 1 kHz can be found in Appendix H.

B. Linewidth of the generated VUV laser

We argue that the linewidth condition given by Eq. (6) can be satisfied in the FWM process. Previously, it has

been extensively studied and shown [64–67] that HHG processes in gases are governed by the coherent temporal dynamics of the electron. Advances in VUV frequency-comb technology [31,68,69] have demonstrated the capability for generating VUV radiation with coherence time greater than 1 s, allowing for subhertz spectral resolution for HHG processes. Particularly interesting for the current work is the third-harmonic generation [31], which also relies on the third-order nonlinearity, and is a special case of FWM process when $\omega_1 = \omega_2 = \omega_3$. These results support our assumption that the cw FWM process will also preserve good phase coherence. Assuming that both the 750.3 and 710.1 nm lasers have equal uncorrelated phase noise, the VUV linewidth scales as $17 \times \Gamma_f/2\pi$, with $\Gamma_f/2\pi$ denoting the linewidth of the fundamental beams. This implies that fundamental linewidths less than 59 Hz are sufficient to satisfy condition Eq. (6), a value well within reach using lasers locked to ultrastable high-finesse cavities [70,71].

V. CONCLUSIONS

In conclusion, we propose using four-wave mixing in cadmium vapor to generate continuous-wave laser light for precision spectroscopy of the ^{229}Th isomeric transition. Based on state-of-the-art *ab initio* electron-correlation calculations, we find that cadmium has large transition-matrix elements, leading to a significant third-order susceptibility near the isomeric transition wavelength for a resonance-enhanced four-wave mixing scheme. With incident laser powers of 3 W at 375.2 nm and 6 W at 710.1 nm, under the optimal phase-matching conditions, more than $30\ \mu\text{W}$ of VUV light at 148.4 nm can be produced. The VUV power can be further enhanced by coupling the incident laser beams into a high-finesse cavity, thereby increasing the effective circulating power in the nonlinear medium [72–74]. The resulting intense VUV laser with a sub-kHz linewidth will enable precision spectroscopy of the ^{229}Th isomeric transition, driving single-ion nuclear Rabi oscillations in an ion trap [21,75,76], observing collective effects of nuclear transitions in thorium-doped crystals [77], and constructing a nuclear optical clock [16,17].

Moreover, using cadmium as a nonlinear medium, we conservatively predict efficient cw VUV generation over a broad range from 140 to 176 nm with commercially available high-power lasers. This wavelength coverage is relevant to a range of applications, including laser cooling and state detection of Al^+ ions and high-resolution atomic and molecular spectroscopy. Finally, applying the same calculation method to other group-12 elements such as zinc and mercury suggests that they may also serve as potential candidates for generating 148.4-nm VUV light. Other divalent elements, e.g., magnesium, are natural targets for future investigation within this framework.

After completion of this work, we experimentally realized the proposed approach [78], providing direct support for the scheme's feasibility and the accuracy of the present analysis.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: 5^1D_2 AS THE TWO-PHOTON RESONANCE STATE

We have also performed calculations for the case with 5^1D_2 as the two-photon resonance state. For $\omega_1 = \omega_2$, the incident lasers are 337.7 and 1223.4 nm, as shown in Fig. 4.

The relative contributions to χ_{12} and χ_{34} stemming from different intermediate P states are presented in Fig. 5.

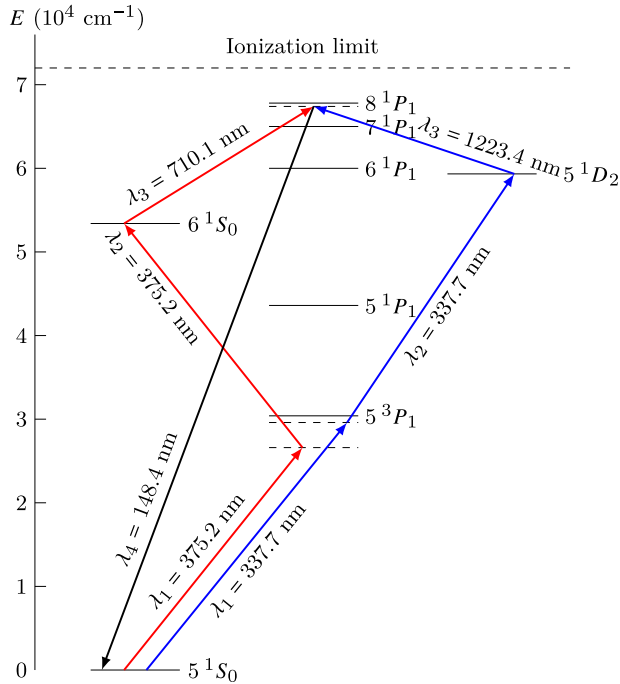


FIG. 4. Simplified energy levels of neutral cadmium in terms of wave number. The red and blue arrows represent the incident lasers λ_1 , λ_2 , λ_3 with 6^1S_0 and 5^1D_2 as two-photon resonance states, respectively. The black arrow indicates the generated VUV laser at 148.4 nm.

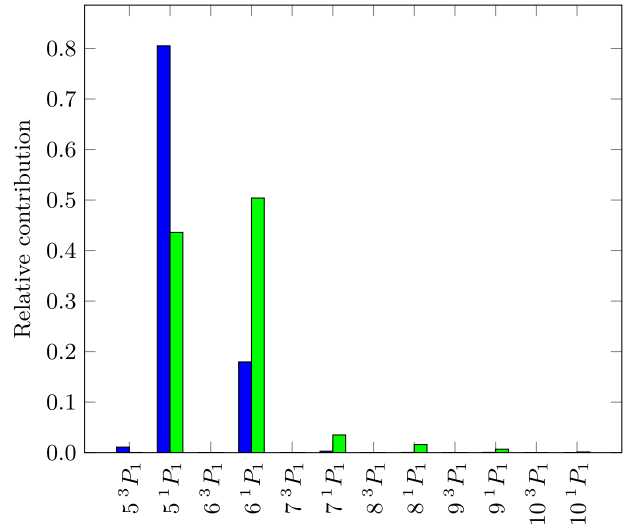


FIG. 5. The normalized relative contributions to χ_{12} (blue bars) and χ_{34} (green bars) for the scheme exploiting the 5^1D_2 as the two-photon resonance state from a set of intermediate P states up to 10^1P_1 .

Similar to the case with 6^1S_0 as the two-photon resonance state, the coupling to the 5^1P_1 and 6^1P_1 states dominates the nonlinear susceptibility, and contributions from all triplet states and states above 8^1P_1 are negligible.

The calculated phase-matching function $G(bN\Delta k_a)$, the nonlinear susceptibility $\chi_a^{(3)}$, and the generated VUV power P_4 in the vicinity of ^{229}Th isomeric transition in the case of pure ^{114}Cd isotope are shown in Fig. 6. The optimal phase-matching temperature is $T = 570^\circ\text{C}$ for $b = 1.5$ mm at 148.4 nm. With the same laser powers (3 W at 337.7 nm and 6 W at 1223.4 nm), around half of the VUV power can be generated compared to the case exploiting 6^1S_0 as the two-photon resonance state. Nevertheless, the power of the commercially available 1223.4 nm laser can be as high as 30 W [79], making the 5^1D_2 scheme also appealing.

APPENDIX B: PHASE MATCHING

It can be shown that the wave-vector mismatch $\Delta k_a = (k_4 - k_1 - k_2 - k_3)/N$ is independent of the number density of the nonlinear medium N . The wave vector k_i is related to the refractive index $n(\omega_i)$ by

$$k_i = n(\omega_i)\omega_i/c, \quad (\text{B1})$$

where [80]

$$n(\omega_i) \simeq 1 + \frac{1}{2}\text{Re}[\chi^{(1)}] = 1 + \frac{Ne^2}{2\epsilon_0 m_e} \sum_m \frac{f_{mg}}{\omega_{mg}^2 - \omega_i^2}. \quad (\text{B2})$$

Here f_{mg} and ω_{mg} are the oscillator strength and transition frequency, respectively. The oscillator strength f_{mg} is given

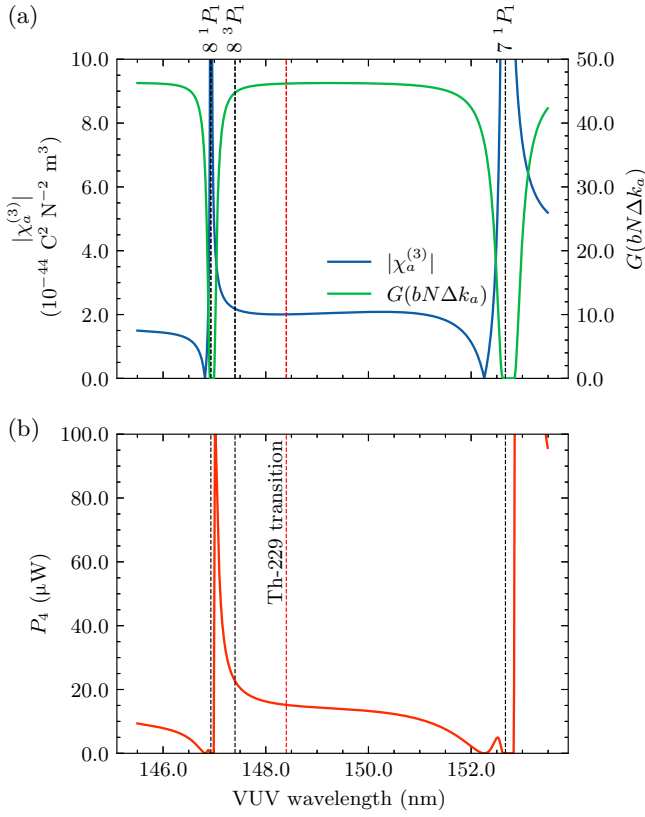


FIG. 6. Calculated (a) nonlinear susceptibility $|\chi_a^{(3)}|$, phase-matching function $G(bN\Delta k_a)$, and (b) VUV yield P_4 in the vicinity of ^{229}Th isomeric transition with the 5^1D_2 as the two-photon resonance state. The confocal parameter is $b = 1.5 \text{ mm}$, the temperature is $T = 570^\circ\text{C}$, and the power of the incident beams is 3 W for the 337.7-nm beam and 6 W for the 1223.4-nm beam. The VUV wavelength range (145.5–153.5 nm) on the horizontal axis corresponds to varying λ_3 from 1051.7 to 1687.2 nm.

by [80]

$$f_{mg} = \frac{2m\omega_{mg}|\mu_{mg}|^2}{3\hbar e^2}, \quad (\text{B3})$$

with the dipole-moment matrix element μ_{mg} related to its z component μ_{mg} by Wigner-Eckart Theorem [44].

By making use of expression $\omega_1 + \omega_2 + \omega_3 = \omega_4$, we get

$$\Delta k_a = \frac{e^2}{2\epsilon_0 m_e c} \sum_m \left(\frac{f_{mg}\omega_4}{\omega_{mg}^2 - \omega_4^2} - \sum_{j=1,2,3} \frac{f_{mg}\omega_j}{\omega_{mg}^2 - \omega_j^2} \right). \quad (\text{B4})$$

Therefore, Δk_a does not depend on the number density of the nonlinear medium, but the frequencies of incident and generated laser beams.

The phase-matching function in the tight focus limit can be written explicitly as [49]

$$G(bN\Delta k_a) = G(b\Delta k) = \begin{cases} 0 & \text{for } b\Delta k > 0, \\ \pi^2 (b\Delta k)^4 e^{b\Delta k} & \text{for } b\Delta k < 0, \end{cases} \quad (\text{B5})$$

where we define $\Delta k = k_4 - k_1 - k_2 - k_3 = N\Delta k_a$. The function above attains its maximum value at $b\Delta k = -4$.

APPENDIX C: CALCULATION OF $S(\omega_1 + \omega_2)$

The pressure broadening and Doppler broadening are typically much larger than the natural linewidth of the two-photon resonance state, and must be considered for the calculation of $S(\omega_1 + \omega_2)$. The pressure broadening $\Delta\omega_p$ is expressed as

$$\Delta\omega_p = Nd^2 \sqrt{\frac{16\pi k_b T}{m}}, \quad (\text{C1})$$

where d and m are the kinetic diameter and the atomic mass of the nonlinear medium atom, respectively, T the medium temperature. For the calculation of $S(\omega_1 + \omega_2)$, pressure broadening can be directly added to the population decay rate Γ_r by

$$\Gamma'_r = \Gamma_r + 2\Delta\omega_p, \quad (\text{C2})$$

since the decoherence of the collisional dephasing and the damping of the natural decay both contribute to decoherence of the transition. This results in a decrease of the FWM conversion efficiency.

The Doppler broadening is given by

$$\Delta\omega_D = 2\pi\omega_{rg} \sqrt{\frac{8k_b T \ln 2}{mc^2}}. \quad (\text{C3})$$

It can be accounted for by using the Maxwell-Boltzmann distribution to collect contributions from atoms with different velocities [50]

$$S(\omega_1 + \omega_2) = \int_{-\infty}^{+\infty} \frac{1}{\Omega_{rg} - \omega'_1 - \omega'_2} f(v_x) dv_x. \quad (\text{C4})$$

Here $\omega'_i = \omega_i(1 + v_x/c)$ is the angular frequency in the atom frame moving with a longitudinal velocity v_x towards the i th incident beam, and $f(v_x) dv_x$ is the

TABLE I. The values of the field- and mass-shift factors (F and K , respectively) used to extract the isotope shifts (IS) of $5^1S_0 \rightarrow 6^1S_0$ and $5^1S_0 \rightarrow 5^1D_2$ for the calculation of the shape function $S(\omega_1 + \omega_2)$.

Transition	F , MHz/fm ²	K , GHz · u
$5^1S_0 \rightarrow 5^3P_1$	−4069	1583
$5^1S_0 \rightarrow 5^1D_2$	−3838	2295
$5^1S_0 \rightarrow 6^1S_0$	−3022	1926

Maxwell-Boltzmann velocity distribution given by

$$f(v_x) dv_x = \sqrt{\frac{m}{2\pi kT}} \exp\left(-\frac{mv_x^2}{2kT}\right) dv_x. \quad (\text{C5})$$

Substituting Eqs. (C2), (C3), and (C5) into Eq. (C4) and performing nondimensionalization, we get [50]

$$S(\omega_1 + \omega_2) = \frac{1}{w} Z(\zeta). \quad (\text{C6})$$

Here $Z(\zeta)$ is the plasma-dispersion function given by

$$Z(\zeta) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{+\infty} dx \frac{e^{-x^2}}{x - \zeta}, \quad (\text{C7})$$

$$\zeta = \left(\omega_1 + \omega_2 + i \frac{\Gamma_r'}{2} - \omega_{rg} \right) / w, \quad (\text{C8})$$

$$w = \frac{\Delta\omega_D}{2\sqrt{\ln 2}}. \quad (\text{C9})$$

Moving atoms experience detuning from the two-photon resonance condition, and contribute less to the value of $S(\omega_1 + \omega_2)$. This leads to a Doppler-broadening-induced decrease of FWM conversion efficiency.

Note that $S(\omega_1 + \omega_2)$ has a weak correlation with the number density of the nonlinear medium N , since both the pressure broadening and the Doppler broadening depend on the medium temperature, which in turn dictates the number density of the nonlinear medium N . Therefore, the optimal output power requires a phase-matching condition slightly deviating from the optimal phase-matching condition $b\Delta k = -4$ solely determined by optimizing $G(bN\Delta k_a)$.

To account for contributions to $S(\omega_1 + \omega_2)$ from different cadmium isotopes, we calculate the isotope shifts (IS) for the $5^1S_0 - 6^1S_0$ and $5^1S_0 - 5^1D_2$ transitions. Isotope shift consists of two contributions: the field shift, which stems from the nucleus charge distribution, and the mass shift, which is associated with the nuclear recoil effect. For mass numbers A and A' , the isotope shift $\delta\nu^{A',A} = \nu^{A'} - \nu^A$ can be written as

$$\delta\nu^{A',A} = F\delta\langle r^2 \rangle^{A',A} + K \frac{m^A - m^{A'}}{m^{A'}m^A}, \quad (\text{C10})$$

where F is the field shift electronic factor, K the mass shift electronic factor, m^A and $m^{A'}$ the isotope masses, and $\delta\langle r^2 \rangle^{A',A} = \langle r^2 \rangle^{A'} - \langle r^2 \rangle^A$ the difference in the mean-square charge radius of the isotopes. The calculated field-shift and mass-shift factors for the $5^1S_0 - 5^3P_1$, $5^1S_0 - 6^1S_0$, and $5^1S_0 - 5^1D_2$ transitions are presented in Table I. Using the available experimental data on the IS of the

TABLE II. The values of reduced E1 matrix elements for neutral cadmium used in the calculation of the nonlinear susceptibility.

Transition	d , arb. units	Transition	d , arb. units	Transition	d , arb. units
$5^1S_0 \rightarrow 5^3P_1$	0.148	$5^3P_1 \rightarrow 6^1S_0$	0.082	$5^3P_1 \rightarrow 5^1D_2$	0.138
$5^1S_0 \rightarrow 5^1P_1$	3.461	$5^1P_1 \rightarrow 6^1S_0$	4.199	$5^1P_1 \rightarrow 5^1D_2$	5.673
$5^1S_0 \rightarrow 6^3P_1$	0.013	$6^1S_0 \rightarrow 6^3P_1$	0.696	$6^3P_1 \rightarrow 5^1D_2$	0.111
$5^1S_0 \rightarrow 6^1P_1$	0.769	$6^1S_0 \rightarrow 6^1P_1$	7.919	$5^1D_2 \rightarrow 6^1P_1$	12.241
$5^1S_0 \rightarrow 7^3P_1$	0.001	$6^1S_0 \rightarrow 7^3P_1$	0.021	$5^1D_2 \rightarrow 7^3P_1$	0.069
$5^1S_0 \rightarrow 7^1P_1$	0.327	$6^1S_0 \rightarrow 7^1P_1$	1.175	$5^1D_2 \rightarrow 7^1P_1$	0.552
$5^1S_0 \rightarrow 8^3P_1$	0.0004	$6^1S_0 \rightarrow 8^3P_1$	0.007	$5^1D_2 \rightarrow 8^3P_1$	0.034
$5^1S_0 \rightarrow 8^1P_1$	0.196	$6^1S_0 \rightarrow 8^1P_1$	0.441	$5^1D_2 \rightarrow 8^1P_1$	0.149
$5^1S_0 \rightarrow 9^3P_1$	0.0003	$6^1S_0 \rightarrow 9^3P_1$	0.010	$5^1D_2 \rightarrow 9^3P_1$	0.020
$5^1S_0 \rightarrow 9^1P_1$	0.136	$6^1S_0 \rightarrow 9^1P_1$	0.237	$5^1D_2 \rightarrow 9^1P_1$	0.27
$5^1S_0 \rightarrow 10^3P_1$	0.0003	$6^1S_0 \rightarrow 10^3P_1$	0.009	$5^1D_2 \rightarrow 10^3P_1$	0.003
$5^1S_0 \rightarrow 10^1P_1$	0.106	$6^1S_0 \rightarrow 10^1P_1$	0.157	$5^1D_2 \rightarrow 10^1P_1$	0.099
$5^1S_0 \rightarrow 11^3P_1$	0.0002	$6^1S_0 \rightarrow 11^3P_1$	0.006	$5^1D_2 \rightarrow 11^3P_1$	0.007
$5^1S_0 \rightarrow 11^1P_1$	0.081	$6^1S_0 \rightarrow 11^1P_1$	0.112	$5^1D_2 \rightarrow 11^1P_1$	0.038
$5^1S_0 \rightarrow 12^3P_1$	0.0003	$6^1S_0 \rightarrow 12^3P_1$	0.003	$5^1D_2 \rightarrow 12^3P_1$	0.005
$5^1S_0 \rightarrow 12^1P_1$	0.050	$6^1S_0 \rightarrow 12^1P_1$	0.070	$5^1D_2 \rightarrow 12^1P_1$	0.012
$5^1S_0 \rightarrow 13^3P_1$	0.0005	$6^1S_0 \rightarrow 13^3P_1$	0.001	$5^1D_2 \rightarrow 13^3P_1$	0.003
$5^1S_0 \rightarrow 13^1P_1$	0.028	$6^1S_0 \rightarrow 13^1P_1$	0.041	$5^1D_2 \rightarrow 13^1P_1$	0.003
$5^1S_0 \rightarrow 14^3P_1$	0.0005	$6^1S_0 \rightarrow 14^3P_1$	0.0006	$5^1D_2 \rightarrow 14^3P_1$	0.002
$5^1S_0 \rightarrow 14^1P_1$	0.017	$6^1S_0 \rightarrow 14^1P_1$	0.027	$5^1D_2 \rightarrow 14^1P_1$	0.001

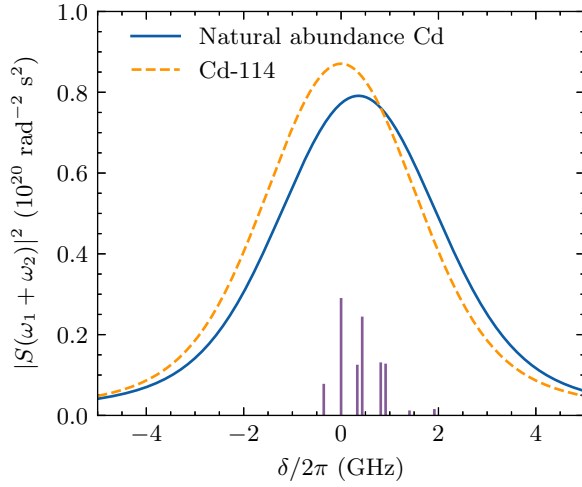


FIG. 7. Calculated $|S(\omega_1 + \omega_2)|^2$ for cadmium with natural abundance (dashed orange line), and for the pure ^{114}Cd isotope (solid blue line). $\delta/2\pi$ denotes the detuning from the 6^1S_0 state of the pure ^{114}Cd isotope. The vertical purple lines show the isotope shifts and the relative abundance.

$5^1S_0 - 5^3P_1$ transition, we extract $\delta \langle r^2 \rangle^{A',A}$ and use it to calculate the IS of the $5^1S_0 - 6^1S_0$ and $5^1S_0 - 5^1D_2$ transitions. The comparison of the shape function $S(\omega_1 + \omega_2)$ between pure ^{114}Cd and cadmium with natural abundance is shown in Fig. 7, where the pure ^{114}Cd isotope gives 5% more output power.

APPENDIX D: SATURATION AND ABSORPTION EFFECTS

In the calculation of the VUV power according to Eq. (1) in the main text, we assume the small signal limit, which is only valid when saturation and linear absorption effects are negligible. Saturation is defined as the deviation from the relation $P_4 \propto P_1 P_2 P_3$, which holds only when the intensities of the incident and generated beams are low.

At high incident laser intensities, two-photon resonance Raman-type absorption and resonantly enhanced ionization become significant, pumping the population from the 5^1S_0 ground state to the 6^1S_0 state and the ionization continuum, respectively. The depletion of the ground-state population reduces the third-order nonlinear susceptibility $\chi_a^{(3)}$, thereby decreasing the output power. On the other hand, if the population in the 6^1S_0 state becomes sufficiently high to form a population inversion between the 6^1S_0 and the 5^1P_1 states, amplified spontaneous emission (ASE) may occur. ASE broadens the 6^1S_0 state, reducing the $S(\omega_1 + \omega_2)$ factor and the output power. Additionally, the altered population distribution changes the refractive index n , potentially disrupting the phase-matching condition.

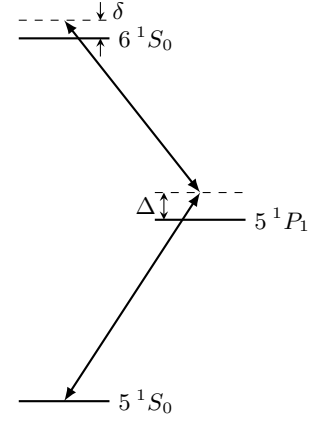


FIG. 8. $5^1S_0 - 6^1S_0$ Raman-type transition.

Other saturation mechanisms, such as absorption and refractive index changes induced by third-order and higher-order processes, as well as pump depletion arising from competing processes, are estimated to be negligible in our case [81].

1. Population introduced by two-photon absorption

We adiabatically eliminate the intermediate states, and reduce the three-level system to a two level system (see Fig. 8). The two-photon Rabi frequency is

$$\Omega = \frac{\Omega_1 \Omega_2}{2\Delta}, \quad (\text{D1})$$

where $\Omega_{1,2}$ are the single-photon Rabi frequencies, and Δ is the detuning from the 5^1P_1 state. We neglect the coupling through other intermediate P states.

The population of the two-photon resonance state 6^1S_0 is estimated as [82]

$$\rho_{ee} = \frac{\Omega^2/2}{\Omega^2 + \frac{2\Gamma_r}{\Gamma_r'}(\delta^2 + \Gamma_r'^2/4)}, \quad (\text{D2})$$

where $\delta = \omega'_1 + \omega'_2 - \omega_{rg}$ is the two-photon detuning, $\omega'_i = \omega_i(1 + v_x/c)$ is the frequency seen in the moving atom frame, Γ_r is the total population decay rate and Γ_r' includes the pressure broadening effect.

The depletion effect of the ground-state population ρ_{gg} on $S(\omega_1 + \omega_2)$ can be introduced as

$$S'(\omega_1 + \omega_2) = \int_{-\infty}^{+\infty} \frac{1}{\Omega_{rg} - \omega'_1 - \omega'_2} \rho_{gg} f(v_x) dv_x. \quad (\text{D3})$$

The $|S'(\omega_1 + \omega_2)|^2$ is dented due to two-photon resonance Raman-type absorption (see Fig. 9). This decrease can be mitigated by slightly detuning from the exact two-photon resonance condition.

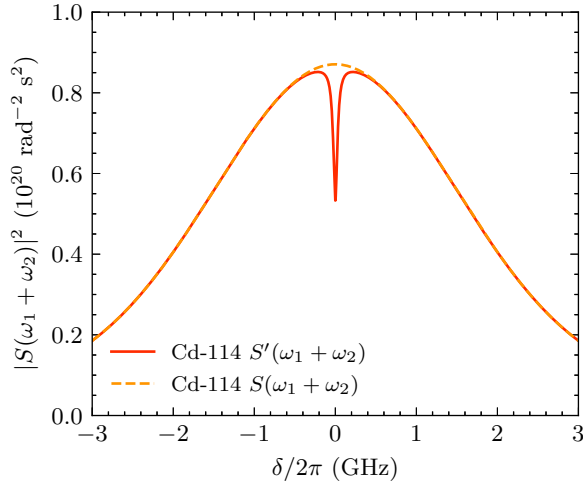


FIG. 9. Calculated $|S(\omega_1 + \omega_2)|^2$ with (solid red line) and without (dashed orange line) considering the depletion effect of the ground-state population due to the two-photon absorption. $\delta/2\pi$ denotes the detuning from the 6^1S_0 state of the pure ^{114}Cd isotope. We assume using the pure ^{114}Cd isotope.

2. Amplified spontaneous emission (ASE) effects

The population inversion between the 6^1S_0 state and the lower-lying $5^{1,3}P_1$ states can cause amplified spontaneous emission. This is another possible source of saturation, which can be represented by a diagram in Fig. 10.

For relatively small laser intensities, the ASE broadening can be accounted for by replacing the Doppler width in Eq. (C6) by [50]

$$\Delta\omega_D \rightarrow (\Delta\omega_D^2 + \Delta\omega_{\text{ASE}}^2)^{1/2}, \quad (\text{D4})$$

where $\Delta\omega_{\text{ASE}}$ is the power-broadened width associated with the ASE.

To estimate the possible broadening caused by the ASE process for the 6^1S_0 state, we follow a derivation in Ref. [83]. For a Lorentzian line, the ASE intensity is

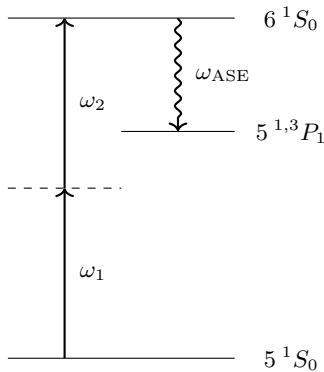


FIG. 10. Amplified spontaneous emission process.

given by

$$I_{\text{ASE}} = \phi I_s \left(\frac{\Omega}{4\pi} \right) \frac{(G-1)^{3/2}}{(G \ln G)^{1/2}}. \quad (\text{D5})$$

Here I_s is the saturation intensity, ϕ the fluorescence quantum yield, Ω the emission solid angle, and G the gain of the active medium. We assume the active medium length to be twice the Rayleigh range, and use the parameters in the last section to calculate the two-photon Rabi frequency. The power-broadened width associated with the ASE is estimated [84] to be $\Delta\omega_{\text{ASE}} \approx 25$ MHz, which is negligible compared to the Doppler width.

3. Linear absorption of the beams

Linear absorption is typically negligible. However, if any of the waves are close to the resonant states, or if the nonlinear medium vapor zone is very long, linear absorption should be considered. The saturated vapor pressure of cadmium as a function of temperature is shown in Fig. 12.

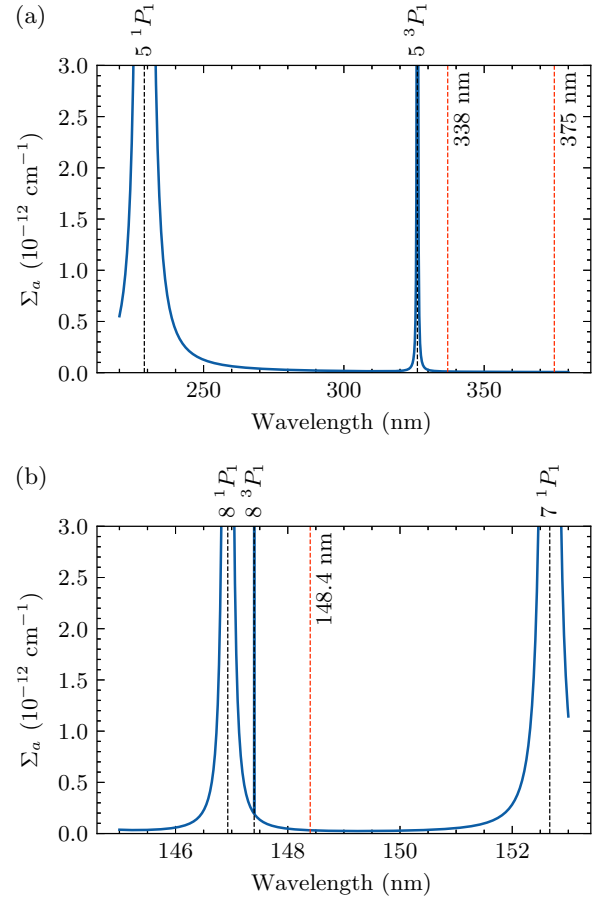


FIG. 11. Absorption cross section (in cm^{-1}) in the wavelength vicinity of (a) input beams, (b) the output VUV beam, calculated for $N = 7.2 \times 10^{17} \text{ cm}^{-3}$, or $T = 580^\circ\text{C}$.

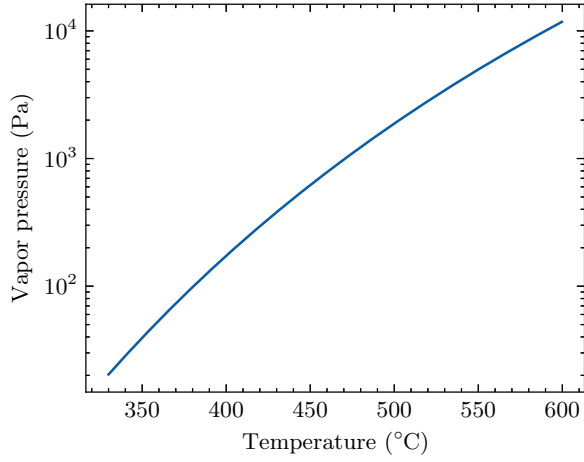


FIG. 12. Cadmium vapor pressure. Typically, we get 8500 Pa of cadmium at 580 °C.

The calculated absorption cross section for different wavelengths is plotted in Fig. 11. The absorption cross section is negligible at 375.2 and 148.4 nm.

4. Resonantly enhanced ionization

The main channels of resonantly enhanced ionization are as follows: (i) cadmium atoms in the ground state (5^1S_0) simultaneously absorbing three 375.2-nm photons to reach the ionization continuum, and (ii) cadmium atoms in the 6^1S_0 excited state absorbing one additional 375.2-nm photon to the continuum [85].

The first process can be estimated through cadmium absorption rate of three 375.2-nm photons to the ionization continuum [51]

$$R_{ng}^{(3)} = \bar{\sigma}_{ng}^{(3)} \bar{I}^3, \quad (D6)$$

where $R_{ng}^{(3)}$ is the transition rate for three-photon absorption, $\bar{\sigma}_{ng}^{(3)}$ the three-photon absorption cross section and $\bar{I}$ the photon flux of the 375.2-nm laser.

In an order-of-magnitude estimation of $R_{ng}^{(3)}$ under the two-photon resonance condition, $\bar{\sigma}_{ng}^{(3)}$ is approximated as [51]

$$\bar{\sigma}_{ng}^{(3)} \approx \frac{|\mu_{nr}\mu_{rm}\mu_{mg}|^2}{8\epsilon_0^3 \hbar^3 c^3 \Gamma_r^2}, \quad (D7)$$

where $\mu_{mg,rm,nr}$ are the dipole-moment matrix elements with g representing the 5^1S_0 ground state, m the allowed intermediate states, namely $m^1,3P_1$ states, r the 6^1S_0 state, and n the ionization continuum. Γ_r is the population decay rate of the 6^1S_0 state of cadmium. Assuming $\mu_{mg} = \mu_{rm} = \mu_{nr} = ea_0 = 8 \times 10^{-30}$ C m and $\Gamma_r = 2\pi \times 1$ MHz, $R_{ng}^{(3)}$ at the laser focus is approximately 10^6 s $^{-1}$ for a 3-W 375.2-nm beam with a 1.5-mm confocal parameter.

The second process corresponds to one-photon absorption from the 6^1S_0 state to the continuum,

$$R_{nr}^{(1)} = \bar{\sigma}_{nr}^{(1)}(\omega) \bar{I}, \quad (D8)$$

where, $\bar{\sigma}_{nr}^{(1)} = (3\lambda^2/2\pi)\rho_{ee}$ is the linear absorption cross-section with λ the wavelength of the 375.2 nm light, and ρ_{ee} the excited-state population. Considering $\rho_{ee} = 0.1$, the $R_{nr}^{(1)}$ is calculated approximately 4×10^5 s $^{-1}$.

Meanwhile, cadmium atoms enter and leave the laser interaction region on a timescale $t \sim N_c \lambda_m / v \approx 5 \times 10^{-8}$ s, where the mean free path is $\lambda_m = 1/(\sqrt{2}n\sigma) \approx 5$ μ m, $N_c = (w_0/\lambda_m)^2 \sim 4$ is the average number of collisions for a 3D random walk across the beam of waist $w_0 \approx 10$ μ m, and $v \approx 4 \times 10^2$ m/s is the mean atomic speed. This transit time is much shorter than the ionization timescale, ensuring rapid replenishment of any ionized cadmium atoms [86].

Finally, since the applied laser fields are far detuned from the $n^1,3P_1$ resonances, population accumulation in these states is negligible, and ionization channels originating from them can be safely ignored.

APPENDIX E: NOBLE GAS FOR PHASE MATCHING

At around 148.4 nm, cadmium is negatively dispersive, meaning the refractive index is smaller at smaller wavelengths. If the required confocal parameter b for a high-output VUV power is too small to be practical, one could exploit the positive dispersion of noble gases for wave-vector mismatch compensation [50]. This extra compensation can also be employed to loosen the focus (i.e., enlarge b) such that the saturation effect due to the high light intensity in the focus can be mitigated. Our calculations show that krypton and xenon gases are good choices for this purpose. For the confocal parameter $b = 1.5$ mm adopted in the main text, adding about 0.5 bar of krypton gas could improve the VUV power from 30 to 50 μ W.

APPENDIX F: WHEN $\omega_1 \neq \omega_2$

In the more general case where $\omega_1 \neq \omega_2$, the third-order nonlinear susceptibility $\chi_a^{(3)}$ can be improved by a few orders of magnitude by tuning ω_1 close to the $5^1,3P_1$ resonance states while keeping the two-photon resonance condition ($\omega_1 + \omega_2$ remains unchanged) [48]. However, the strong dispersion experienced by ω_1 in the vicinity of these resonance states spoils the phase matching condition. Therefore, the gain in $\chi_a^{(3)}$ does not translate into a net increase in the VUV output power at 148.4 nm without additional phase-mismatch compensation (e.g., using noble gases).

By contrast, for generating VUV wavelengths close to higher-lying $n^1,3P_1$ resonances (e.g., around 167 nm), the

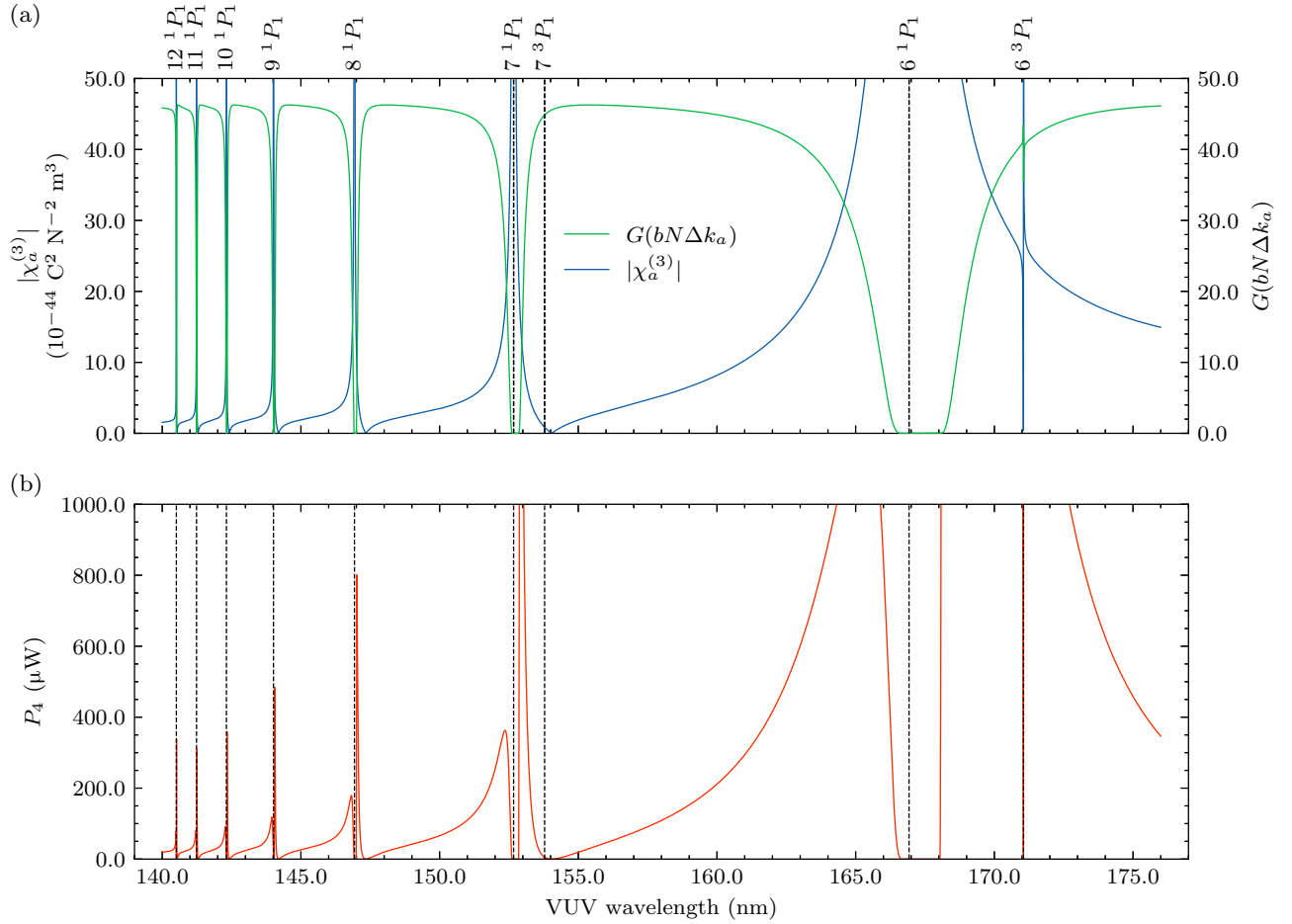


FIG. 13. Calculated (a) nonlinear susceptibility $|\chi_a^{(3)}|$, phase-matching function $G(bN\Delta k_a)$, and (b) VUV yield P_4 from 140 to 176 nm, with the 6^1S_0 as the two-photon resonance state. The confocal parameter is $b = 1.5$ mm, the temperature is $T = 580^\circ\text{C}$, and the power of the incident beams is 3 W for the 375.2-nm beam and 6 W for the varying $\lambda_3 = 552 \sim 2851$ nm beam. We omit the label for the n^3P_1 states for n higher than 7.

dispersion of ω_1 can be exploited to counteract that of ω_4 and thus restore phase matching [48].

APPENDIX G: VUV GENERATION FROM 140 TO 176 NM

VUV generation using cadmium is predicted to be efficient over the 140 to 176 nm range (see Fig. 13). Resonances with stronger transition-matrix elements, e.g., the 6^1P_1 state, enhance the third-order nonlinear susceptibility $\chi_a^{(3)}$ and thus increase the VUV power in their vicinity. The power nulls that appear near the n^1P_1 resonances arise from destructive phase matching. As discussed in Appendix F, these minima can be mitigated by tuning ω_1 close to the $5^{1,3}P_1$ resonances while maintaining the two-photon resonance condition, thereby extending the VUV generation range toward the strong resonant lines.

APPENDIX H: THEORY OF THE OPTICAL NUCLEAR EXCITATION

We assume the VUV laser with a power of $10 \mu\text{W}$ is focused to a spot diameter of $1.8 \mu\text{m}$. To observe the nuclear Rabi oscillation, the VUV laser linewidth needs to satisfy

$$\Gamma_l < 2\pi \times 1 \text{ kHz}. \quad (\text{H1})$$

The interaction of the ^{229}Th nucleus with the VUV light has been treated in Ref. [63]. For simplicity, we consider a nuclear two-level system consisting of a ground state g and an excited state e . The laser-induced Rabi frequency Ω_{eg} can be written as

$$\Omega_{eg} = \sqrt{\frac{2\pi c^2 I_l \Gamma_\gamma}{\hbar \omega_0^3}}, \quad (\text{H2})$$

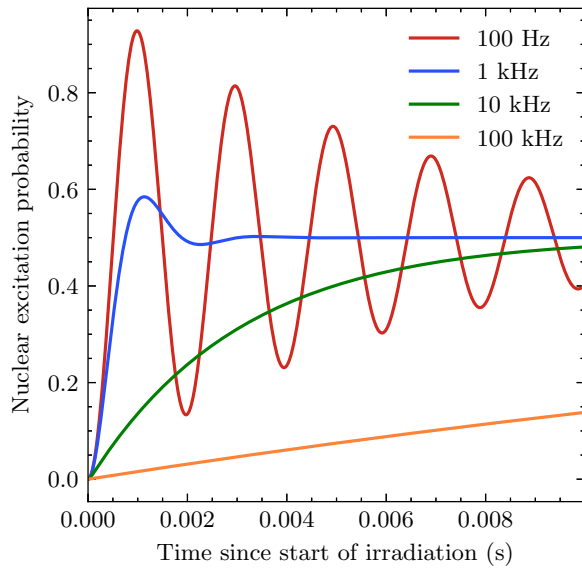


FIG. 14. The time-dependent nuclear excitation probability for a single ^{229}Th nucleus as a function of resonant irradiation time for different laser linewidths.

where I_l is the laser intensity, ω_0 the angular frequency of the nuclear transition, and Γ_γ the radiative decay rate of the nuclear excited state.

For an extremely low excited-state decay rate, as in the case of the ^{229}Th isomeric transition, the condition for the VUV laser linewidth $\Gamma_l/2\pi$ to observe the Rabi oscillation can be written as

$$\Gamma_l < 2\Omega_{eg} = 2\sqrt{\frac{2\pi c^2 I_l \Gamma_\gamma}{\hbar \omega_0^3}}. \quad (\text{H3})$$

The nuclear excitation probability as a function of irradiation time for different VUV laser linewidths is plotted in Fig. 14. As expected, we observe the Rabi oscillation when the VUV laser linewidth is close to or below 1 kHz.

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