PAPER

A spectrally bright wavelength-switchable vacuum ultraviolet source driven by quantum coherence in strong-field-ionized molecules

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Abstract

We report generation of spectrally bright vacuum ultraviolet (VUV) and deep UV (DUV) coherent radiations at the wavelengths of 192 nm, 198 nm and 204 nm. These DUV/VUV radiations originate from resonant four-wave mixing assisted by quantum coherence in tunnel-ionized CO molecules. The electronic coherence is created when the pump pulses resonantly excite electronic transitions of CO⁺. The technique allows for the selective switch of DUV/VUV wavelengths provided by the abundant energy levels of molecular ions. The developed source can have arbitrary polarization states by manipulating the polarization of the pump pulse. It also exhibits a spectral bandwidth of 5~7 cm⁻¹, a divergence angle of 3~5 mrad, a pulse duration of ∼10 ps, and a photon flux of ∼10¹⁰ photons/s. The superior temporal and spectral properties give rise to a broadband Raman comb in the DUV/VUV region.

1. Introduction

Coherent vacuum ultraviolet (VUV, 100–200 nm) and deep UV (DUV, 200–300 nm) light sources have been widely applied in numerous studies, such as biochemical kinetics [1, 2], surface structure analysis [3, 4], precision measurements [5, 6], atmospheric chemistry [7, 8], and nanolithography [9]. Free-electron lasers have been built to provide wavelength-tunable short-wavelength light sources with high intensity, good coherence, short pulse duration as well as well-defined polarization [10, 11]. However, these large facilities are expensive and typically of large footprint sizes, which limit the widespread applications. Recently, table-top DUV/VUV sources have been developed based on nonlinear optical crystals of KBe₂BO₃F₂ family. The shortest wavelength reported so far is 149.8 nm using this approach as determined by the absorption edge of the crystal [12]. In comparison with solids, gaseous media usually have a broader transparent window, a higher damage threshold as well as a lower dispersion, facilitating the extension of coherent radiation toward shorter wavelengths. One has generated DUV/VUV radiations through four-wave mixing (FWM), harmonic generation from neutral gas [13, 14], or the dispersive wave in the gas-filled fiber [15, 16].

It is also of vital importance to flexibly control the parameters of the table-top DUV/VUV coherent light sources such as the wavelength, bandwidth, pulse duration and polarization. For instance, the polarization manipulation of DUV/VUV sources is a key prerequisite for studying the optical properties of chiral species [17] and magnetic textures [18, 19], which cannot be readily achieved by harmonic generation in gas or...
nonlinear crystals due to the sensitivity of nonlinear polarization or phase matching to polarization of the driver laser. The temporal or spectral shaping of DUV/VUV radiation is critical to precision spectroscopy [20], laser cooling [21], imaging and metrology [22], which remains a challenge owing to the lack of optics in this regime. These difficulties motivate us to explore an alternative solution, allowing for the table-top generation and flexible control of DUV/VUV sources.

Here, we demonstrate a table-top DUV/VUV coherent light source around 200 nm with unique characteristics including narrow bandwidth, picosecond pulse duration, switchable wavelength, and fully controllable polarization. Such DUV/VUV coherent radiations are generated through resonant FWM in the strong-field-ionized CO molecules. We choose CO$^+$ as the nonlinear medium due to the following reasons. On the one hand, it has suitable electronic-state intervals and large transition dipole moments [23], allowing for resonant excitation with a femtosecond laser available in most ultrafast laboratories and generation of coherent radiations in the DUV/VUV region. On the other hand, its abundant vibrational quantum states enable generation of multi-wavelength-switchable DUV/VUV radiations. Owing to the creation of electronic coherence during resonant excitation, the developed DUV/VUV source has the sufficiently high spectral resolution and relatively high photon flux, and does not require the time synchronization of two pump fields. In addition, the polarization of the generated radiation, including ellipticity, helicity and main-axis direction, can be controlled by changing the polarization of the pump laser.

More importantly, the generated DUV/VUV radiation with the picosecond duration originates from nonlinear frequency conversion from the femtosecond pump pulses. The combination of the pump pulses and the DUV/VUV radiation shows great advantages in coherent Raman scattering, because it allows us to utilize both high spectral resolution with the DUV/VUV source and impulsive preparation of rotational coherence with the femtosecond pump laser. Furthermore, DUV/VUV Raman spectroscopy has higher efficiency [24] and smaller fluorescence interference [25] than common Raman spectroscopy with the infrared or visible light. Based on these advantages, we show the generation of DUV/VUV Raman comb with more than 100 Raman sidebands. The developed source will open promising applications in biological identification, explosives sensing, gas-phase thermometry, etc.

2. Experimental details

The experiment was performed with a commercial Ti:sapphire laser (Legend Elite-Duo, Coherent, Inc.), which delivers near-infrared (NIR) laser pulses with the 800 nm wavelength, 40 fs duration, ~6 mJ per pulse energy, and 1 kHz repetition rate. The experimental setup is composed of three parts, i.e. generation of DUV/VUV radiation, generation of Raman comb and cross-correlation measurement, as shown in figure 1. The experimental details in each part are described as follows.

2.1. Generation of DUV/VUV coherent radiations

The laser pulse from Ti:sapphire laser was divided into three beams. Beam 1 with the energy of 3.8 mJ served as the NIR pump pulse. Beam 2 with the energy of 2 mJ was converted as the wavelength-tunable violet pulse. NIR and violet pulses were used for the pump sources of DUV/VUV coherent radiations. Beam 3 with the energy of 0.2 mJ was utilized for the temporal-domain measurement of DUV/VUV radiation and Raman comb. The violet pump pulse was generated by the double frequency of beam 2 in the BBO1. Its wavelength can be tuned from 384 nm to 423 nm by adjusting phase-matched angle. A bandpass filter was used in the violet beam to obtain a near-Gaussian spectrum with a bandwidth of ~3 nm. The corresponding pulse duration is evaluated to be ~100 fs in the Fourier transform limit. The violet pump power was tuned using a continuously variable attenuator. The two beams were combined by DM2 and then were collinearly focused into 10 mbar CO gas using an $f = 60$ cm lens (L3). The combination of an $f = 25$ cm lens (L1) and an $f = -10$ cm lens (L2) was used to compensate for the chromatic aberration of the focal lens (L3) as well as to enhance the efficiency of frequency doubling. The time delay between NIR and violet pump pulses was controlled by a motorized translation stage (delay 1). Unless otherwise specified, their polarizations were set parallel to each other and the delay 1 was chosen at 1.26 ps. The DUV/VUV radiation exiting from the CO gas chamber was spectrally filtered and then was completely collected into a spectrometer (Shamrock 500i, Andor) using a plano-convex lens for spectral analysis.

The polarization of DUV radiation was controlled by tuning the ellipticity of the violet pump pulse, which was realized by the combination of HWP2 and QWP. Such a design allows us to continuously change the ellipticity by rotating HWP2 while remaining the main axis unchanged. The polarization state of DUV radiation was measured with a Rochon prism, and was recorded by the spectrometer. The polarization sensitivity of the spectrometer has been calibrated.
2.2. Generation of Raman comb

DUV/VUV Raman comb was produced by the residual NIR pulse and the generated DUV radiation at 204 nm. The O$_2$ gas was chosen as the Raman-active medium due to its large Raman cross section and favorable rotational constant [26]. The NIR and DUV beams exited from CO gas were collimated by an $f = 50$ cm concave mirror (CM1), and then collinearly focused into O$_2$ gas by an $f = 100$ cm concave mirror (CM2). In the Raman scattering region, the time delay between NIR and DUV pulses is about 10 ps. The NIR femtosecond laser was used to impulsively excite rotational coherence of O$_2$ molecules. When the 204 nm radiation travels through the coherently excited O$_2$ molecules, it will be scattered to generate Raman comb in the DUV/VUV region. After spectral filtering, Raman signal was collected into a spectrometer for spectral analysis.

2.3. Cross-correlation measurement

Temporal information of DUV/VUV Raman comb was characterized by the cross-correlation measurement. The Raman signal generated in 200 mbar O$_2$ gas together with the residual 204 nm radiation were combined with beam 3, and then were launched into BBO2 to generate difference-frequency signal (DFS). The spectrum of DFS was recorded at different time delays between beam 3 and Raman comb. The delay was controlled with a motorized translation stage (delay II). The zero delay is defined as the moment when the NIR pulse for rotational excitation of O$_2$ molecules arrives the Raman scattering region. The increasing delay means that beam 3 is sent later. From the time-frequency diagram obtained by cross-correlation measurement, we can approximately obtain the temporal envelopes of DUV radiation at 204 nm and Raman comb.

3. Results and discussion

3.1. Generation of DUV/VUV coherent radiations

Figure 2(a) shows the energy diagram for generation of DUV/VUV radiations through the resonant interaction of two pump pulses with CO$^+$. We choose three electronic states of CO$^+$, and adopt NIR and violet pump pulses to resonantly excite transitions between these electronic states. The intensity of the NIR pump pulse is sufficiently high to ionize CO molecules. The generated CO$^+$ is mainly populated in $X^2\Sigma^+ (v = 0)$ state. It can be efficiently excited to $A^2\Pi_i (v = 3)$ and $A^2\Pi_i (v = 4)$ states through resonantly absorbing two NIR photons with the wavelength of 803.5 nm and 759.2 nm, respectively. Subsequently, it is further excited to $B^2\Sigma^+ (v = 2, 3, 4)$ states by resonantly absorbing one violet photon. The resonant excitation among these electronic states allows for strong DUV/VUV radiations via resonant FWM of NIR and violet pump pulses. Owing to abundant vibrational energy levels involved in these electronic states, we observed coherent radiations at 204 nm, 198 nm and 192 nm wavelengths by tuning the violet pump wavelength, which correspond to the transition from $B^2\Sigma^+ (v = 2, 3, 4)$ to $X^2\Sigma^+ (v = 0)$, respectively. The typical spectra and spatial profiles of these DUV/VUV radiations are shown in figures 2(b)–(d). All signals exhibit a narrow bandwidth (5~7 cm$^{-1}$), a small divergence angle (3~5 mrad) and a linear polarization.
It should be emphasized that the resonant excitation induces electronic coherence between $X^2\Sigma^+$ and $A^2\Pi_i$ states as well as $A^2\Pi_i$ and $B^2\Sigma^+$ states, which, on the one hand, dramatically enhances DUV/VUV radiations. On the other hand, the quantum coherence between $X^2\Sigma^+$ and $A^2\Pi_i$ states enables FWM when two pump pulses are temporally separated from each other. This is because the macroscopic polarization still exists after the NIR pump pulse, allowing for the subsequent interaction with the delayed violet pulse [27]. As a result, these DUV/VUV radiations show a slow decay when the violet pulse lags behind the NIR pulse. The dynamic evolution of DUV radiation with the delay of two pump pulses will be elaborated later.

We further studied the dependence of DUV/VUV radiations on wavelength and power of the violet pump pulse. Figure 3(a) clearly shows that 192 nm and 198 nm radiations reach the maximum when the violet pump wavelength is tuned to about 390 nm, whereas the 204 nm radiation is the strongest at the pump wavelength of 415 nm. The optimal wavelengths for all radiations are in perfect agreement with that predicted by the resonant FWM process in figure 2(a). Furthermore, 192 nm and 198 nm radiations linearly grow with the increase of violet pump powers, whereas the 204 nm radiation reaches saturation at the higher pump powers, as shown in figure 3(b). The photon flux of the 204 nm radiation is about $4 \times 10^{10}$ photons/s, which is calculated by the measured maximum average power. The strength of the 198 nm signal is comparable to that of the 204 nm signal. In comparison, the 192 nm signal is much weaker than other two signals, which could be attributed to the smaller Franck–Condon factor corresponding to this transition [23] and lower excitation efficiency from $X^2\Sigma^+ (v = 0)$ to $A^2\Pi_i (v = 4)$.

We also examined the evolution of these DUV/VUV signals with the time delay between NIR and violet pump pulses (i.e. delay $\tau$). Unlike the conventional non-resonant FWM which requires a temporal overlap between the interacting waves, these DUV/VUV radiations can still be generated when the NIR and violet pump pulses are temporally separated. When the violet pulse arrives ahead of the NIR pulse (negative delay), these narrow-bandwidth radiations vanish. When the violet pulse lags behind the NIR laser (positive delay), these narrow-bandwidth radiations decay slowly with an increasing delay, as shown in figure 3(c), which reflects the slow decoherence between $X^2\Sigma^+$ and $A^2\Pi_i$ states. Figure 3(c) also shows that these radiations are strongly modulated when changing delay $\tau$. Interestingly, modulation moments match very well with the revival periods of rotational wavepackets of $A^2\Pi_i (v = 3, 4)$ states, which are calculated by $T_{v=3,4} = 1/(2B_{v=3,4}^* c)$ with the rotational constant $B_{v=3}^* = 1.50 \text{ cm}^{-1}$ and $B_{v=4}^* = 1.52 \text{ cm}^{-1}$ [29, 30].
Figure 3. (a) DUV/VUV radiations as a function of the violet pump wavelength. The violet pump power is maintained at 45 mW while changing wavelength. The intensities of three radiations are normalized independently. (b) Dependence of DUV/VUV radiations on the violet pump power. The solid line is the fitting of experimental data. The linear function was used for the fitting for the 192 nm and 198 nm radiation, and the smoothing spline was used for the fitting of the 204 nm radiation. (c) Evolution of DUV/VUV signals with the time delay between NIR and violet pulses. The curves have been vertically shifted for clarity. The error bars in (a) and (b) represent the standard deviation of measurements.

Hence, our scheme for generation of a DUV/VUV source also allows for exploring dynamics of rotational wavepackets of excited states.

3.2. Control of polarization of DUV/VUV coherent radiations

The polarization states of these DUV/VUV coherent radiations can be fully controlled by changing the ellipticity of the violet pump pulse as we show below. For a resonant FWM process $\omega_{\text{FWM}} = 2\omega_1 + \omega_2$ in an isotropic nonlinear medium, the nonlinear polarization is given by \[ P_R = \chi^{(3)R}_{1111} (\omega_{\text{FWM}}; \omega_1, \omega_1, \omega_2) \left\{ 3 (1 - \overline{\eta}) E_1 (E_1 E_2) + 3\overline{\eta} E_2 (E_1 E_2) \right\}. \] (1)

Where $\overline{\eta} = \chi^{(3)R}_{1221}/\chi^{(3)R}_{1111}$ is the depolarization ratio, $\chi^{(3)R}_{1221}$ and $\chi^{(3)R}_{1111}$ are components of the third-order nonlinear susceptibility, and $E_1$ and $E_2$ are electronic fields of NIR and violet pump pulses, respectively. Experimentally, taking the 204 nm radiation as an example, its intensity generated in the case of $E_1 \parallel E_2$ is about 3.4 times of that in the case of $E_1 \perp E_2$. From this measured value, we obtain $\overline{\eta} = 0.54$. For the case of $E_1 = E_1 \hat{y}$, $E_2 = \frac{E_2}{\sqrt{1 + \xi^2}} (\hat{x} + i\xi \hat{y})$, equation (1) becomes

\[ P_R = \frac{3}{\sqrt{1 + \xi^2}} \chi^{(3)R}_{1111} (\omega_{\text{FWM}}; \omega_1, \omega_1, \omega_2) E_1^2 E_2 (0.54\hat{x} + i\xi \hat{y}). \] (2)

Equation (2) clearly shows that we can generate arbitrarily polarized DUV/VUV radiation by simply changing the ellipticity $\xi$ of the violet pump pulse. The main axis of the violet pump pulse is fixed in $\hat{x}$ direction while its ellipticity varies. The polarization state of the 204 nm radiation was measured by recording its spectral intensity as a function of angle of Rochon prism. The measured results are shown in figures 4(a)–(c). We can clearly see that the polarization of the 204 nm radiation can be tuned from nearly linear to circular with the increase of $\xi$. The theoretical ellipticity values for generating linearly, elliptically and circularly polarized DUV radiation in figures 4(a)–(c) are $\xi = 0, 0.27$ and $0.54$ according to equation (2), whereas the corresponding experimental values are $\xi = 0.07, 0.28$ and $0.52$. The deviation between the theoretical and experimental results in figure 4(a) could be caused by the imperfect linear
3.3. Generation of DUV/VUV Raman comb

As demonstrated above, DUV/VUV coherent radiations with narrow bandwidth, selective wavelength and controllable polarization can be produced through the resonant interaction of femtosecond laser pulses with CO$^+$ ions. In this section, we will show the application of the developed DUV/VUV light source in Raman spectroscopy. It should be emphasized that the narrow-bandwidth DUV/VUV radiation obtained by the broadband pump laser has some great advantages in coherent Raman scattering, because it uniquely...
combines the advantages of impulsive preparation of rotational coherence with the femtosecond pump laser and high spectral resolution with the DUV/VUV radiation [33]. In addition, the generated DUV/VUV radiation has the ideal spatial overlap and the proper time delay with the NIR femtosecond pump pulse. These inherent merits facilitate the cascaded Raman scattering to form a DUV/VUV Raman comb. To verify the concept, we studied cascaded rotational Raman scattering in O2 molecules using the residual NIR femtosecond laser and the 204 nm radiation exited from CO gas.

The basic process of cascaded Raman scattering is schematically illustrated in figure 5(a). The NIR femtosecond pump pulse at 800 nm impulsively excites rotational wavepackets of O2 molecules, resulting in time-varying alignment as well as the temporal modulation of optical polarizability of the medium. When the 204 nm probe pulse passes through the modulated medium, its frequency will be shifted, giving rise to the first order Raman scattering. The 1st Raman signal can be further scattered in the coherently excited molecules to generate the 2nd Raman signal. The cascaded process can be extended to higher order due to the initial rotational coherence prepared by the femtosecond pump pulse. In this process, narrow spectral bandwidth of the 204 nm radiation allows us to resolve different rotational quantum states. Since O2 molecules are initially populated in multiple rotational energy levels, each order Raman scattering consists of multiple Raman sidebands. All Raman sidebands from different orders construct a Raman comb.

Figure 5(b) shows anti-Stokes Raman spectra captured at different O2 gas pressures, which consist of many discrete peaks. Their frequency shifts with respect to the 204 nm radiation perfectly match with the calculated values of high-order rotational Raman scattering. As shown in figure 5(a), frequency shifts of the first order Raman scattering obey $\Delta = (4J + 6)B$ with the rotational constant $B = 1.4456 \text{ cm}^{-1}$ of O2 molecules [26]. Because only odd rotational states are populated, Raman sidebands consist of discrete peaks spaced by a constant value of $8B$. Figure 5(b) also indicates that adjacent order Raman scattering is separated by $58B$, which reflects the maximum population in $J = 13$ state at 294 K. We can clearly see that Raman scattering is extended to higher orders with the increase of O2 gas pressure. In the 300 mbar O2 gas, we observed Raman scattering up to 21st order, which forms a Raman comb with more than 100 sidebands. The Raman comb has a quasi-periodic structure with the frequency spacing of $\sim 11.6 \text{ cm}^{-1}$ (i.e. $8B$). It is
noteworthy that such high-order Raman scattering is the result of joint contributions of the broadband pump laser and the narrow-bandwidth DUV radiation, which cannot be readily achieved by other methods.

Owing to the rotational coherence in the impulsively excited O₂ molecules, Raman sidebands in figure 5(b) are phase locked at alignment moments, which corresponds to an ultrashort pulse train in the time domain. To confirm this, we performed a cross-correlation measurement to obtain the temporal information of these Raman signals. The experimental detail is given in the section 2.3. Figure 6(a) shows DFS of the Raman signal together with the residual 204 nm radiation and the 800 nm femtosecond pulse (beam 3) as a function of their relative delay. The measured result clearly indicates that the DUV radiation is modulated at revival moments of rotational wavepackets. The time-frequency diagram is divided into three regions, which approximately reflect the temporal information of anti-Stokes Raman signal, the 204 nm radiation, and Stokes Raman signal, respectively. The temporal envelope of 204 nm radiation is obtained by the spectral integration of DFS in the middle region, and the points at revival moments are excluded. The temporal envelope of Raman comb is obtained by integrating DFS corresponding to the anti-Stokes components. As demonstrated in figure 6(b), the 204 nm radiation shows an asymmetrical temporal envelope with the pulse duration of ~10 ps, whereas the Raman comb corresponds to a femtosecond pulse train with an interval of ~2.88 ps. The interval is equal to one quarter of rotational period $T_{rev}$ of O₂ molecules [26]. The relative intensity of sub-pulses in the pulse train basically follows the temporal envelope of the 204 nm radiation. In principle, the characteristics of the Raman comb such as the wavelength range, frequency spacing and temporal structure can be controlled by adopting different DUV/VUV sources and Raman-active molecules.

4. Conclusion and outlook

To conclude, we have demonstrated generation, wavelength selection, polarization control and application of the DUV/VUV coherent radiations driven by electronic coherence in strong-field-ionized molecules. Specifically speaking, our scheme is based on resonant FWM in molecular ions. In principle, the scheme proposed in this work can be used for constructing picosecond coherent sources in the exotic wavelength ranges such as VUV and infrared regimes. The abundant energy levels of molecular ions provide a large number of operation wavelengths of such sources. These advantages will benefit a broad range of applications ranging from high-resolution spectroscopy and surface science to photochemistry and medicine.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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References

[1] Park S T, Kim S K and Kim M S 2002 Observation of conformation-specific pathways in the photodissociation of 1-iodopropane ions Nature 415 306–8
[2] Cao W, Laurent G, Ben-Itzhak I and Cocke C 2015 Identification of a previously unobserved dissociative ionization pathway in time-resolved photospectroscopy of the deuterium molecule Phys. Rev. Lett. 114 113001
[3] Barnes B, Henn M, Sohn M, Zhou H and Silver R 2019 Assessing form-dependent optical scattering at vacuum- and extreme-ultraviolet wavelengths of nanostructures with two-dimensional periodicity Phys. Rev. Applied 11 064056
[4] Laarmann T et al 2004 Interaction of argon clusters with intense VUV-laser radiation: the role of electronic structure in the energy-deposition process Phys. Rev. Lett. 92 143401

[5] Bergeson S D et al 1998 Measurement of the He ground state lamb shift via the two-photon 1S–2S Transition Phys. Rev. Lett. 80 3475

[6] Dreissen J S, Roth C, Gründeman E L, Krauth E E, Favier M and Eikema K S E 2019 High-precision Ramsey–comb spectroscopy based on high-harmonic generation Phys. Rev. Lett. 123 143001

[7] Caravan R et al 2018 The reaction of hydroxyl and methylperoxy radicals is not a major source of atmospheric methanol Nat. Commun. 9 1–9

[8] Hanna S J, Campuzano-Jost P, Simpson E A, Robb D B, Burak I, Blades M W, Hepburn J W and Bertram A K 2009 A new broadly tunable (7.4–10.2 eV) laser based VUV light source and its first application to solar mass spectrometry Int. J. Mass Spectrom. 279 134–46

[9] Yamada T et al 2016 Nanoparticle chemisorption printing technique for conductive silver patterning with submicron resolution Nat. Commun. 7 11402

[10] Pellegrini C, Marinelli A and Reiche S 2016 The physics of x-ray free-electron lasers Rev. Mod. Phys. 88 015006

[11] Ayvazyan V et al 2002 Generation of UV radiation pulses from a VUV free-electron laser operating in the femtosecond regime Phys. Rev. Lett. 88 104802

[12] Nakazato T, Ito I, Kobayashi Y, Wang X, Chen C and Watanabe S 2016 Phase-matched frequency conversion below 150 nm in KBe2BO3F2 Opt. Express 24 17149–58

[13] Beutler M, Ghofhi M, Noack F and Hertel I V 2010 Generation of sub-50-fs vacuum ultraviolet pulses by four-wave mixing in argon Opt. Lett. 35 1491–3

[14] Galli M et al 2019 Generation of deep ultraviolet sub-2–fs pulses Opt. Lett. 44 1308–11

[15] Köttig F, Tani F, Bierrasch J, Travers J and Russell P 2017 Generation of microjoule pulses in the deep ultraviolet at megahertz repetition rates Optica 4 1272–6

[16] Travers J C, Grigorieva T F, Brahm s C and Bell F 2019 High-energy pulse self-compression and ultraviolet generation through soliton dynamics in hollow capillary fibres Nat. Photon. 13 547–54

[17] Nahon L, García G, Dossmann H, Daly S and Pows I 2010 Effects of dimerization on the photoelectron angular distribution parameters from chiral camphor enantiomers obtained with circularly polarized vacuum-ultraviolet radiation Phys. Rev. A 82 104675–73

[18] Schretter N B M, Pei D, Vergniory M G, Sun Y and Chen Y 2019 Chiral topological semimetal with multifold band crossings and long Fermi arcs Nat. Phys. 15 1–7

[19] Shimojima T et al 2010 Orbital-dependent modifications of electronic structure across the magnetostuctural transition in BaFe2As2 Phys. Rev. Lett. 104 057002

[20] Heeg K P et al 2017 Spectral narrowing of x-ray pulses for precision spectroscopy with nuclear resonances Science 357 375–8

[21] Yamaguchi A, Safronova M, Gibble K and Katori H 2019 Narrow-line cooling and determination of the magic wavelength of Cd Phys. Rev. Lett. 123 113201

[22] Consolino L et al 2017 Spectral purity and tunability of terahertz quantum cascade laser sources based on intracavity difference-frequency generation Sci. Adv. 3 e1603317

[23] Nicholls R W 1962 Franck–Condon factors and r-centroids to high vibrational quantum numbers for three band systems of CO Phys. Rev. 127 3471–5

[24] Nicholls R W 1962 Orbital-dependent modifications of electronic structure across the magnetostuctural transition in BaFe2As2 Phys. Rev. Lett. 104 057002

[25] Asher S and Johnson C 1984 Raman spectroscopy of a coal liquid shows that fluorescence interference is minimized with ultraviolet excitation Science 225 311–3

[26] Fenner W R, Hyatt H A, Kellam J M and Porto S P 1973 Raman cross sections of some simple gases Phil. Trans. R. Soc. Lond. A 262 375–9

[27] Chen J et al 2019 Electronic–coherence-mediated molecular nitrogen–ion lasing in a strong laser field Phys. Rev. A 100 031402(R)

[28] Zhang W et al 2016 Photon energy deposition in strong-field single ionization of multielectron molecules Phys. Rev. Lett. 117 103002

[29] Kepr Kocan A, Ostrowska-Kope M, Piotrowska-Domagała I and Zachwieja M 2004 New spectroscopic studies of the Comet–Tail (A2Π – X2Σ+) system of the CO+ molecule J. Mol. Spectrosc. 228 665–75

[30] Mulliken R S and Robert S 1931 The interpretation of band spectra. Part I Ic. Empirical band types Rev. Mod. Phys. 3 89–155

[31] Akmkanov S, Bunkin A, Ivanov S and Koroteev N 1978 Polarization active Raman spectroscopy and coherent Raman ellipsometry Sov. Phys. JETP 47 667–77

[32] Boyd R W 2008 Nonlinear Optics (Amsterdam: Elsevier)

[33] Liu Z et al 2020 Extremely nonlinear Raman interaction of an ultrashort nitrogen ion laser with an impulsively excited molecular wave packet Phys. Rev. A 101 043404