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Application No. 23 209 230.4 - 1001	Ref. HJS01350EP	Date 13.03.2025
Applicant Hefei National Laboratory, et al		

Communication under Rule 71(3) EPC

1. Intention to grant

You are informed that the examining division intends to grant a European patent on the basis of the above application, with the text and drawings and the related bibliographic data as indicated below

A copy of the relevant documents is enclosed

1.1 Text intended for grant

In the text for the Contracting States:

AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC ME MK MT NL NO
PL PT RO RS SE SI SK SM TR

Description, Pages

1-12 filed in electronic form on 04-06-2024

Claims, Numbers

1-8 filed in electronic form on 04-06-2024

Drawings, Sheets

1/3-3/3 as originally filed

With the following amendments to the above-mentioned documents proposed by the division

Description, Pages 1, 12

Claims, Numbers 1

Comments

DESCRIPTION

Page 1: Deletion of an incorporation-by-reference not essential to the invention (EPC Guidelines F-III, 8)

Page 12: Deletion of a statement which implies that the extent of protection may be expanded in some vague and not precisely defined way (EPC Guidelines F-IV, 4.4)

CLAIMS

Page 1, Claim 1: Absent or incorrect two-part form of claim (Rule 43(1) EPC)

See also the comments in enclosed EPO Form 2906.

1.2 Bibliographic data

The title of the invention in the three official languages of the European Patent Office, the international patent classification, the designated contracting states, the registered name(s) of the applicant(s) and the other bibliographic data are shown on **EPO Form 2056** (enclosed).

2 Invitation

You are invited, **within a non-extendable period of four months** of notification of this communication,

2.1 to EITHER approve the text communicated above and verify the bibliographic data (Rule 71(5) EPC)

- (1) by filing a translation of the claim(s) in the other two official languages of the EPO

	Fee code	EUR
(2a) by paying the fee for grant including the fee for publication minus any amount already paid (Rule 71a(5) EPC):	007	1080.00 0.00
Total amount:		1080.00
(3) by paying additional claims fees under Rule 71(4) EPC; number of claims fees payable: 0 minus any amount already paid (Rule 71a(5) EPC):	016	0.00 0.00
Total amount:		0.00

Important: If the translations of the claims and fees have already been filed and paid respectively in reply to a previous communication under Rule 71(3) EPC, e.g. in the case of resumption of examination after approval (see Guidelines C-V, 6), **agreement as to the text to be granted** (Rule 71a(1) EPC) must be expressed within the same time limit (e.g. by approving the text and verifying the bibliographic data, by confirming that grant proceedings can go ahead with the documents on file and/or by stating which translations of the claims already on file are to be used).

Note 1: See "Notes concerning fee payments" below.

Note 2: Any overpaid "minus" amounts will be refunded when the decision to grant (EPO Form 2006A) has been issued.

Note 3: For the calculation of the grant fee under Article 2(2), No. 7, RFees (old fee structure), the number of pages is determined on the basis of a clean copy of the application documents, in which text deleted as a result of any amendments by the examining division is not shown.
Such clean copy is made available via on-line file inspection only.

2.2 OR, in the case of disapproval, to request reasoned amendments or corrections to the text communicated above or keep to the latest text submitted by you (Rule 71(6) EPC).

In this case the translations of the claims and fee payments mentioned under point 2.1 above are NOT due.

The terms "amendment(s)" and "correction(s)" refer only to amendments or corrections of the application documents and not of other documents (e.g. bibliographic data, the designation of the inventor, etc.).

If filing amendments, you must identify them and indicate the basis for them in the application as filed. Failure to meet either requirement may lead to a communication from the examining division requesting that you correct this deficiency (Rule 137(4) EPC).

2.3 Bibliographic data

Where you request a change or correction of bibliographic data in response to the Rule 71(3) communication, this will **not** cause the sending of a further communication under Rule 71(3) EPC. You will still have to pay the fees and file translations in reply to the Rule 71(3) communication in the case of 2.1 above, unless you also file a reasoned request for amendments or corrections in response to the Rule 71(3) communication (see case 2.2 above).

3. Loss of rights

If neither of the two possible actions above (see points 2.1 or 2.2) is performed in due time, the European patent application will be deemed to be withdrawn (Rule 71(7) EPC).

4. Further procedure

4.1 In the case of point 2.1 above

4.1.1 The decision to grant the European patent will be issued, and the **mention of the grant** of the patent will be published in the European Patent Bulletin, if the requirements concerning the translation of the claims and the payment of all fees are fulfilled and there is agreement as to the text to be granted (Rule 71a(1) EPC).

Note on payment of the renewal fee:

If a renewal fee becomes due before the next possible date for publication of the mention of the grant of the European patent, publication will be effected only after the renewal fee and any additional fee have been paid (Rule 71a(4) EPC).

Under Article 86(2) EPC, the obligation to pay renewal fees to the European Patent Office terminates with the payment of the renewal fee due in respect of the year in which the mention of the grant of the European patent is published.

Note on payment of the designation fee(s):

If the designation fee(s) become(s) due after the communication under Rule 71(3) EPC, the mention of the grant of the European patent will not be published until these fees have been paid (Rule 71a(3) EPC).

4.1.2 After publication, the **European patent specification** can be downloaded free of charge from the EPO publication server <https://data.epo.org/publication-server>.

4.1.3 Filing of translations in the contracting states

As regards translation requirements prescribed by the contracting states under Article 65(1) EPC, please consult the website of the European Patent Office

www.epo.org → Law & practice → Legal texts, National law relating to the EPC

www.epo.org → Law & practice → All Legal texts → London Agreement

In the case of a valid extension or validation

As regards translation requirements prescribed by the extension or validation states, please consult the website of the European Patent Office

www.epo.org → Law & practice → Legal texts, National law relating to the EPC

Failure to supply a prescribed translation in a contracting state, or in an extension or validation state may result in the patent being deemed to be void *ab initio* in the state concerned (Art. 65(3) EPC).

4.2 In the case of 2.2 above

If the present communication under Rule 71(3) EPC is based on an auxiliary request and, within the time limit, you maintain the main request or a higher ranking request which was not found allowable, the application may be refused (Art. 97(2) EPC).

If the examining division gives its consent to the requested amendments or corrections, it will issue a new communication under Rule 71(3) EPC; otherwise, it shall resume the examination proceedings (Rule 71(6) EPC).

5. Filing of a divisional application

Any divisional application relating to this European patent application must be filed directly with the European Patent Office in Munich, The Hague or Berlin and will be in the language of the proceedings for the present application, or if the latter was not in an official language of the EPO, the divisional application may be filed in the language of the present application as filed (see Article 76(1) and Rule 36(2) EPC). Any such divisional application must be filed while the present application is still pending (Rule 36(1) EPC; Guidelines A-IV, 1.1.1).

6. Notes concerning fee payments

6.1 Making payments

For payments made via deposit account, please note that as from 1 December 2017 debit orders will only be carried out if filed in an electronically processable format (xml), using an accepted means of filing as laid down in the Arrangements for deposit accounts (ADA), published in the Supplementary publication in the Official Journal.

All relevant information related to the modes of payment of fees to the EPO can be retrieved from the EPO website at "**Making Payments**".

6.2 Information concerning fee amounts

Procedural fees are usually adjusted every two years, on even years, with effect from 1 April. Therefore, before making a payment, parties should verify the amounts actually due on the date of payment using the applicable version of the Schedule of fees and expenses, published as a Supplement to the Official Journal of the EPO, available on the EPO website (www.epo.org) at www.epo.org/schedule-of-fees. The "Schedule of fees" table allows the viewing, downloading and searching of individual fee amounts, both current and previous.

6.3 Note to users of the automatic debiting procedure

The fee for grant, including the fee for publication, and any additional claims fees due under Rule 71(4) EPC will be debited automatically on the date of filing of the translations of the claims, or on the last day of the period of this communication. However, if the designation fee(s) become(s) due as set out in Rule 71a(3) EPC and/or a renewal fee becomes due as set out in Rule 71a(4) EPC, these should be paid separately by another permitted way of payment in order not to delay the publication of the mention of the grant. The same applies in these circumstances to the payment of extension and validation fees.

Examining Division:

Chair: Sauerer, Christof
2nd member: Huenges, Alexandra
1st member: Politsch, Erich

**Annexes:**

Applicants not using the Mailbox can access patent literature via Espacenet
Text intended for grant
EPO Form 2056
EPO Form 2906

Annex to EPO Form 2004, Communication pursuant to Rule 71(3) EPC

Bibliographical data of European patent application No. 23 209 230.4

For the intended grant of the European patent, the bibliographical data are set out below, for information:

Title of invention: - VORRICHTUNG ZUR QUANTITATIVEN DETEKTION VON ISOTOPOLOGEN VON KOHLENDIOXID MITTELS DUAL-PHOTONEN-ABSORPTION UND SPEKTROMETER
- APPARATUS FOR QUANTITATIVELY DETECTING ISOTOPOLOGUE OF CARBON DIOXIDE USING DUAL-PHOTON ABSORPTION AND SPECTROMETER
- APPAREIL DE DÉTECTION QUANTITATIVE D'ISOTOPOLOGUE DE DIOXYDE DE CARBONE UTILISANT UN SPECTROMÈTRE ET UN ABSORPTION À DEUX PHOTONS

Classification: INV. G01N21/39 G01J3/42 G01J3/433 ADD. G01N21/03 G01N21/3504 G01N33/00

Date of filing: 10.11.2023

Priority claimed: CN / 24.11.2022 / CNA202211479911

Contracting States*
for which fees have been paid: AttributeDTO(name=DesignatedStateCountryCodeCurrent, objectID=null, value=AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC ME MK MT NL NO PL PT RO RS SE SI SK SM TR, type=StringAttribute)

Extension States*
for which fees have been paid:

Validation States*
for which fees have been paid:

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*) If the time limit for the payment of designation fees according to Rule 39(1) EPC has not yet expired and the applicant has not withdrawn any designation, **all Contracting States/Extension**

States/Validation States are currently still deemed to be designated. See also Rule 71a(3) EPC and, if applicable, the above Note to users of the automatic debiting procedure.

**) If two or more applicants have designated different Contracting States, this is indicated here.

Datum
Date 13.03.2025
Date

Blatt
Sheet 1
Feuille




Anmelde-Nr:
Application No: 23 209 230.4
Demande n°:

National prior rights within the meaning of Article 139(2) EPC are not a bar to the grant of a European patent in proceedings before the EPO. Therefore, the EPO is not required to search for and assess such rights (see EPC Guidelines, H-III, 4.4). Applicants may, however, consider the procedural option under Rule 138 EPC in view of the effects of such rights in national proceedings and/or before the Unified Patent Court (Article 3 Regulation (EU) No 1257/2012). As a support service free of charge for the applicant in this context, the applicant is hereby offered non-binding information on a search for and prima facie relevance assessment of national prior rights by the examining division. It is the applicant's responsibility to assess such national prior rights and any use of the procedural option under Rule 138 EPC (see EPC Guidelines, H-III, 4.4).

The applicant is informed that no prima facie relevant national prior rights were found.

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**APPARATUS FOR QUANTITATIVELY DETECTING ISOTOPOLOGUE OF
CARBON DIOXIDE USING DUAL-PHOTON ABSORPTION AND
SPECTROMETER**

5 [0001] The present application claims priority to Chinese Patent Application No. 202211479911.8, titled "APPARATUS FOR QUANTITATIVELY DETECTING ISOTOPOLOGUE OF CARBON DIOXIDE USING DUAL-PHOTON ABSORPTION AND SPECTROMETER ", filed on November 24, 2022 with the China National Intellectual Property  Administration.  

~~Administration, which is incorporated herein by reference in its entirety.~~

FIELD

[0002] The present disclosure relates to the technical field of spectral detection, and in particular to a device, which is for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption, and a spectrometer.

BACKGROUND

[0003] Carbon dioxide is one of the key greenhouse gases in the earth atmosphere and a key factor in the carbon cycle. Due to the isotope fractionation, content of each isotopologue in a carbon dioxide sample may be quantitatively detected to distinguish a source of the sample, and hence the quantitative detection is widely used in fields such as chronometric dating, tracing, forensic identification, environmental testing, and drug metabolism. Laser spectroscopy is one of important means at present for detecting isotopes in carbon dioxide, and hence has a great potential in the quantitative detection. A main concept of sensitive detection on isotopes in carbon dioxide through the laser spectroscopy is detecting a spectral
20
25 signal of an isotopologue of carbon dioxide through cavity ring-down spectroscopy.

[0004] At present, determination of isotopologues in carbon dioxide through the laser spectroscopy requires higher sensitivity and is subject to apparently insufficient selectivity. Moreover, the laser spectroscopy is limited by the Doppler broadening on spectra linewidth, and hence an isotopologue of carbon dioxide cannot be effectively distinguished from an

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isotopologue of another carbon compound or another molecule. Hence, it is difficult to achieve accurate quantitative measurement. As an example, "Detection of Radiocarbon Dioxide with Double-Resonance Absorption Spectroscopy" by Yan-dong Tan, et al. (Chinese Journal of Chemical Physics, vol. 34, No. 4, page 373-380) and "Seeded optical parametric oscillator light source for precision spectroscopy" by Zhang Z. -T. et al. (Optics Letters, vol. 45, No. 4, page 1013) discloses a method and a system for detecting radiocarbon dioxide with double-resonance absorption spectroscopy. As another example, "Optical Measurement of Radiocarbon below Unity Fraction Modern by Linear Absorption Spectroscopy" by Fleisher Adam J. et al. discloses a system for measuring radiocarbon by using linear absorption spectroscopy, in which a conventional distributed feedback quantum cascade laser serves as a laser source. As another example, patent document with publication No. US 2018/052047 A1 discloses a carbon isotope analyser, in which a laser beam incident on an optical resonator containing analyte gases yields absorption spectra.

SUMMARY

[0004a] The invention is defined in the appended claims.

[0005] In view of the above, an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption and a spectrometer are provided to address at least the above issue.

[0006] In a first aspect, an apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption are provided according to an embodiment of the present disclosure. The apparatus comprises a laser source, a laser frequency stabilizer, a sample chamber, a signal detector, and a signal analyzer, where: the sample chamber comprises an optical resonator and a piezoelectric ceramic; the laser source is configured to output a laser beam; the laser frequency stabilizer is configured to lock the laser beam to a mode frequency of the optical resonator; the piezoelectric ceramic is configured to adjust a length of the optical resonator to alter the mode frequency of the optical resonator to match energy levels of a target molecular isotopologue; the signal detector is configured to detect a transmission intensity of the light beam passing the optical resonator to obtain a dual-photon absorption signal; and the signal analyzer is configured to analyze and process the dual-photon absorption signal to obtain a concentration of the target molecular isotopologue.

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[0007] In an embodiment, the laser beam is a continuous infrared laser beam, and a power of the laser beam is greater than 100mW.

[0008] In an embodiment, a fineness of the optical resonator is higher than 60000.

[0009] The energy levels of the target molecular isotopologue comprise: energy levels of dual-photon absorption, where an energy level representing single-photon absorption is between the energy levels of dual-photon absorption.

[0010] Energy of a photon having the mode frequency matching the energy levels of the target molecular isotopologue is equal to a half of an energy difference between the energy levels of the target molecular isotopologue.

[0011] In an embodiment, fluctuations of temperature of the optical resonator are less than 10mK.

[0012] In an embodiment, the sample chamber further comprises a temperature controller, where the temperature controller is configured to control temperature of the optical resonator.

[0013] In an embodiment, the signal detector comprises a detector and a signal amplifier which are integrated, where the detector is configured to detect the transmission intensity of the light beam passing the optical resonator to obtain the dual-photon absorption signal, and the signal amplifier is configured to amplify the obtained dual-photon absorption signal.

[0014] In an embodiment, the signal analyzer is configured to: invoke a signal processing program to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue, and display the concentration of the target molecular isotopologue via an interactive interface.

[0015] In a second aspect, a spectrometer is further provided according to an embodiment of the present disclosure. The spectrometer comprises the apparatus in any foregoing embodiment.

[0016] In comparison with conventional technology, embodiments of the present disclosure achieves at least following beneficial effects.

[0017] The apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption are provided, comprising the laser source, the laser frequency stabilizer, the sample chamber, the signal detector, and the signal analyzer. The sample

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chamber comprises the optical resonator and the piezoelectric ceramic. The laser source is configured to output the laser beam. The laser frequency stabilizer is configured to lock the laser beam to the mode frequency of the optical resonator. The piezoelectric ceramic is configured to adjust the length of the optical resonator to alter the mode frequency of the optical resonator to match energy levels of the target molecular isotopologue. The signal detector is configured to detect the transmission intensity of the light beam passing the optical resonator to obtain the dual-photon absorption signal. The signal analyzer is configured to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue. Herein the apparatus operates based on dual-photon absorptiometry.

The laser frequency stabilizer controls a frequency of the laser beam to lock the laser beam onto the optical resonator, so that the frequency of the laser beam is consistent with the mode frequency of the optical resonator. The length of the optical resonator is adjusted through the piezoelectric ceramic to tune the mode frequency of the optical resonator, and thereby tune the frequency of the laser beam towards a frequency matching the energy levels of the target molecular isotopologue. The target molecular isotopologue is selectively excited once the frequency of the laser beam matches the energy levels of the target molecular isotopologue. The transmission intensity of the laser beam passing the optical resonator is measured via the signal detector. When an intensity of the laser beam is strong enough, the target molecular isotopologue is capable to absorb two photons, which is reflected in the dual-photon absorption signal. The signal analyzer is configured to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue. A spectral linewidth of the dual-photon absorption is narrow due to its Doppler-free characteristic, which can effectively improve a spectral resolution and selectivity on the isotopologue. Hence, an isotopologue of carbon dioxide can be effectively distinguished in spectra from isotopologues of other carbon compounds and isotopologues of other molecules. Since amplitude of the dual-photon absorption signal is proportional to an intensity of the laser beam outputted from the laser source, the dual-photon absorption signal having sufficient strength can be obtained through increasing the intensity of the transmission light. Accordingly, sensitive quantitative detection of isotopologues of carbon dioxide can be achieved.

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BRIEF DESCRIPTION OF THE DRAWINGS

[0018] In order to explain embodiments of the present disclosure or the technical solutions in the conventional technology more clearly, the drawings needed to be used in the description of the embodiments or the conventional technology will be briefly introduced below.

5 Obviously, the drawings in the following description are only embodiments of the present disclosure. For those of ordinary skill in the art, other drawings can be obtained based on the provided drawings without creative efforts.

[0019] Figure 1 is a schematic structural diagram of an apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption according to an
10 embodiment of the present disclosure.

[0020] Figure 2 is a diagram of energy-level matching for an apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption according to an embodiment of the present disclosure.

[0021] Figure 3 is a schematic structural diagram of an apparatus for quantitatively
15 detecting isotopologue of carbon dioxide using dual-photon absorption according to another embodiment of the present disclosure.

[0022] Figure 4 is a schematic structural diagram of an apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption according to another embodiment of the present disclosure.

20 [0023] Figure 5 is a schematic structural diagram of an apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption according to another embodiment of the present disclosure.

[0024] Figure 6 is a schematic diagram of a structure of a spectrometer according to an
25 embodiment of the present disclosure.

DETAILED DESCRIPTION OF EMBODIMENTS

[0025] The technical solutions in the embodiments of the present disclosure will be clearly and completely described below with reference to the accompanying drawings in the embodiments of the present disclosure. Obviously, the described embodiments are only

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some of the embodiments of the present disclosure, rather than all the embodiments. Based on the embodiments of the present disclosure, all other embodiments obtained by those of ordinary skill in the art without creative efforts fall within the scope of protection of the present disclosure.

5 **[0026]** In order to clarify and facilitate understanding of the above objects, features and advantages of the present disclosure, hereinafter the present disclosure is further illustrated in detail in conjunction of the drawings and specific embodiments.

[0027] An apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption is provided according to an embodiment of the present disclosure.

10 Reference is made to Figure 1, which is a schematic structural diagram of an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption according to an embodiment of the present disclosure. As shown in Figure 1, the apparatus comprises: a laser source 1, a laser frequency stabilizer 2, a sample chamber 3, a signal detector 4, and a signal analyzer 5. The sample chamber 3 comprises an optical resonator 6
15 and a piezoelectric ceramic 7.

[0028] The laser source 1 is configured to output a laser beam. The laser frequency stabilizer 2 is configured to lock the laser beam to a mode frequency of the optical resonator 6.

[0029] In one embodiment, an optical path of the laser beam outputted by the laser source 1
20 may be deflected via a reflector 8, such that the laser beam is reflected into a beam splitter 9. A part of the laser beam goes into the sample chamber 3 from the beam splitter 9, and another part of the laser beam goes into the laser frequency stabilizer 2. The laser frequency stabilizer 2 may modulate and demodulate a frequency and a phase of the laser beam going into the laser frequency stabilizer 2. The laser frequency stabilizer 2 generates a signal
25 representing an error, generates another signal as a negative feedback of the error, and transmits the negative-feedback signal to the laser source 1, so as to control a frequency of the laser beam emitted by the laser source 1 to be identical to a mode frequency of the optical resonator 6. In addition, the laser frequency stabilizer 2 may further determine automatically
30 whether the laser beam output by the laser source 1 is locked to the mode frequency of the optical resonator 6. In case of negative determination, the laser frequency stabilizer 2 may

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control the frequency of the laser beam outputted by the laser source 1 automatically, so as to lock the laser beam to the mode frequency of the optical resonator 6.

[0030] The piezoelectric ceramic 7 is configured to adjust a length of the optical resonator 6, so as to alter the mode frequency of the optical resonator 6 and thereby match the frequency of the laser beam with energy levels of the target molecular isotopologue.

[0031] The signal detector 4 is configured to detect a transmission intensity of the laser beam passing the optical resonator 6, so as to obtain a dual-photon absorption signal.

[0032] The signal analyzer 5 is configured to analyze and process the dual-photon absorption signal to obtain a concentration of the target molecular isotopologue.

[0033] The apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption are provided, comprising the laser source 1, the laser frequency stabilizer 2, the sample chamber 3, the signal detector 4, and the signal analyzer 5. The sample chamber 3 comprises the optical resonator 6 and the piezoelectric ceramic 7. The laser source 1 is configured to output the laser beam. The laser frequency stabilizer 2 is configured to lock the laser beam to the mode frequency of the optical resonator 6. The piezoelectric ceramic 7 is configured to adjust the length of the optical resonator 6 to alter the mode frequency of the optical resonator 6 to match energy levels of the target molecular isotopologue. The signal detector 4 is configured to detect the transmission intensity of the light beam passing the optical resonator 6 to obtain the dual-photon absorption signal. The signal analyzer 5 is configured to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue. Herein the apparatus operates based on dual-photon absorptiometry. The laser frequency stabilizer 2 controls a frequency of the laser beam to lock the laser beam onto the optical resonator 6, so that the frequency of the laser beam is consistent with the mode frequency of the optical resonator 6. The length of the optical resonator 6 is adjusted through the piezoelectric ceramic 7 to tune the mode frequency of the optical resonator 6, and thereby tune the frequency of the laser beam towards a frequency matching the energy levels of the target molecular isotopologue. The target molecular isotopologue is selectively excited once the frequency of the laser beam matches the energy levels of the target molecular isotopologue. The transmission intensity of the laser beam passing the optical resonator 6 is measured via the signal detector 4. When an

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intensity of the laser beam is strong enough, the target molecular isotopologue is capable to absorb two photons, which is reflected in the dual-photon absorption signal. The signal analyzer 5 is configured to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue. A spectral linewidth of the dual-photon absorption is narrow due to its Doppler-free characteristic, which can effectively improve a spectral resolution and selectivity on the isotopologue. Hence, an isotopologue of carbon dioxide can be effectively distinguished in spectra from isotopologues of other carbon compounds and isotopologues of other molecules. Since amplitude of the dual-photon absorption signal is proportional to a square of an intensity of transmission light after the excitation, the dual-photon absorption signal having sufficient strength can be obtained through increasing the intensity of the transmission light. Accordingly, sensitive quantitative detection of isotopologues of carbon dioxide can be achieved.

[0034] Hereinafter a structure of the foregoing apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption is further illustrated in details in conjunction with optional embodiments.

[0035] In an embodiment, the laser source 1 is an infrared laser source. The laser beam is a continuous infrared laser beam, and a power of the laser beam may be greater than 100mW.

[0036] In an embodiment, the optical resonator 6 is a high-fineness optical resonator. For example, fineness of the optical resonator 6 is higher than 60,000.

[0037] In an embodiment, a wavelength of the laser beam outputted by the laser source 1 can be rapidly tuned. A tunable bandwidth of the wavelength of the laser beam may be 1MHz preferably, and the fineness of the optical resonator 6 for a single laser frequency is higher than 60,000.

[0038] In an embodiment, the energy levels of the target molecular isotopologue comprise energy levels of dual-photon absorption. An energy level representing single-photon absorption is between the energy levels of dual-photon absorption.

[0039] In an embodiment, the mode frequency matching the energy levels of the target molecular isotopologue refers to that energy of a photon having the mode frequency is equal to, or substantially equal to, a half of an energy difference between the energy levels of the target molecular isotopologue.

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[0040] In an embodiment, the energy levels of the target molecular isotopologue are energy levels which are selected. Reference is made to Figure 2, which is a diagram of energy-level matching for an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption according to an embodiment of the present disclosure. In Figure 2, E1 and E3 denote the energy levels of dual-photon absorption, and E2 denotes an energy level representing (i.e., relating to) single-photon absorption. E2 is located approximately at the middle between E1 and E3, that is, the single-photon absorption takes places under energy approximately half of energy required for the dual-photon absorption. A difference between the energy level representing single-photon absorption and a right middle position between the energy levels of dual-photon absorption does not exceed δ . Moreover, when the frequency of the laser beam outputted by the laser source 1 is controlled to be a frequency corresponding a half of the energy difference between the energy levels of the target molecular isotopologue, the frequency of the laser beam is close to the energy level representing single-photon absorption. In such case, strength of the dual-photon absorption signal can be greatly improved.

[0041] Hereinafter a structure of the sample chamber 3 in the foregoing apparatus for quantitatively detecting the isotopologue of carbon dioxide using dual-photon absorption is further illustrated in conjunction with optional embodiments. Reference is made to Figure 3, which illustrates a schematic structural diagram of an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption according to another embodiment. As shown in Figure 3, the sample chamber 3 further comprises a temperature controller 10, which is configured to control temperature of the optical resonator 6.

[0042] In an embodiment, fluctuations of the temperature of the optical resonator 6 are within 10mK.

[0043] In an embodiment, a means of keeping the fluctuations of the temperature of the optical resonator 6 within 10 mK includes but is not limited to controlling the optical resonator 6 through the temperature controller 10 K. Other means of temperature adjustment may also feasible for controlling the temperature of the optical resonator 6 to fluctuate within 10mK.

[0044] Hereinafter a structure of the signal detector 4 in the foregoing apparatus for

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quantitatively detecting the isotopologue of carbon dioxide using dual-photon absorption is further illustrated in conjunction with optional embodiments. Reference is made to Figure 4, which illustrates a schematic structural diagram of an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption according to another
5 embodiment. As shown in Figure 4, the signal detector 4 comprises a detector 11 and a signal amplifier 12 which are integrated.

[0045] The detector 11 may be a sensitive detector. In an embodiment, the detector is configured to detect a transmission intensity of the laser beam passing the optical resonator 6, so as to obtain the dual-photon absorption signal.

10 **[0046]** The signal amplifier 12 may be a low-noise signal amplifier. In an embodiment, the signal amplifier 12 is configured to amplify the obtained dual-photon absorption signal.

[0047] In an embodiment, the target molecular isotopologue can be selectively excited when the frequency of the laser beam outputted by the laser source 1 matches the energy levels of the target molecular isotopologue, and the light beam passes the optical resonator 6
15 as a transmitting beam after the target molecular isotopologue is excited and then is detected by the detector 11. Thereby, the dual-photon absorption signal is obtained. Since the dual-photon absorption signal detected by the signal detector 4 is weak, the obtained dual-photon absorption signal is first amplified by the low-noise signal amplifier 12, which facilitates subsequent processing of the dual-photon absorption signal.

20 **[0048]** Hereinafter a structure of the signal analyzer 5 in the foregoing apparatus for quantitatively detecting the isotopologue of carbon dioxide using dual-photon absorption is further illustrated in conjunction with optional embodiments. Reference is made to Figure 5, which illustrates a schematic structural diagram of an apparatus for quantitatively detecting an isotopologue of carbon dioxide using dual-photon absorption according to another
25 embodiment. As shown in Figure 5, the signal analyzer 5 may comprise a storage medium storing a signal processing program 13, and provide an interactive interface 14. The signal analyzer 5 is configured to invoke the signal processing program 13 to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue, and display the concentration of the target molecular isotopologue via an
30 interactive interface 14.

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[0049] In an embodiment, the interactive interface 14 may update the displayed concentration of the target molecular isotopologue in real time, and operations may be performed on the displayed concentration of the target molecular isotopologue via the interactive interface 14. The interactive interface 14 comprises but is not limited to an electronic device such as a computer and a mobile phone.

[0050] On a basis of the foregoing embodiments, a spectrometer is further provided according to an embodiment of the present disclosure. Reference is made to Figure 6, which is a schematic diagram of a spectrometer according to an embodiment of the present disclosure. The spectrometer comprises the apparatus for quantitatively detecting the isotopologue of carbon dioxide by the dual-photon absorption as described in any foregoing embodiment.

[0051] In some embodiments, the spectrometer has the same features as the apparatuses in the foregoing embodiments.

[0052] In addition, it is noted that the apparatus and the spectrometer as described in the foregoing embodiments of the present disclosure is not limited to quantitative detection on isotopologue(s) of carbon dioxide, and may be utilized to detect isotopologue(s) of another molecule.

[0053] Hereinabove the apparatus for quantitatively detecting the isotopologue of carbon dioxide by dual-photon absorption and the spectrometer are illustrated in detail according to embodiments of the present disclosure. Specific examples are applied herein to illustrate the principles and embodiments of the present disclosure. Description of the above embodiments is only intended for helping understand technical solutions and a core concept of the present disclosure. Those of ordinary skill in the art can make changes in specific implementations and application scopes according to the concept of the present disclosure. In summary, content of this specification should not be construed as a limitation of the present disclosure.

[0054] The embodiments of the present disclosure are described in a progressive manner, and each embodiment places emphasis on the difference from other embodiments. Therefore, one embodiment can refer to other embodiments for the same or similar parts. Since the spectrometer disclosed in the embodiments corresponds to the apparatus disclosed in the

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embodiments, the description of the spectrometer is simple, and reference may be made to the relevant part of the apparatus.

[0055] It should be noted that, the relationship terms such as "first", "second" and the like are only used herein to distinguish one entity or operation from another, rather than to necessitate or imply that an actual relationship or order exists between the entities or operations. Furthermore, the terms such as "include", "comprise" or any other variants thereof means to be non-exclusive. Therefore, a process, a method, an article or a device including a series of elements include not only the disclosed elements but also other elements that are not clearly enumerated, or further include inherent elements of the process, the method, the article or the device. Unless expressively limited, the statement "including a..." does not exclude the case that other similar elements may exist in the process, the method, the article or the device other than enumerated elements.

[0056] According to the description of the disclosed embodiments, those skilled in the art can implement or use the present disclosure. Various modifications made to these embodiments may be obvious to those skilled in the art, and the general principle defined herein may be implemented in other embodiments without departing from the ~~spirit or~~ scope of the present disclosure. Therefore, the present disclosure is not limited to the embodiments described herein but ~~is~~ **is** only limited by the appended claims. ~~is~~ **is**

~~conforms to a widest scope in accordance with principles and novel features disclosed in the present disclosure.~~

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CLAIMS

1. An apparatus for quantitatively detecting isotopologue of carbon dioxide using dual-photon absorption, comprising:

a laser source (1), a laser frequency stabilizer (2), a sample chamber (3), a signal detector (4), and a signal analyzer (5), wherein:

the sample chamber (3) comprises an optical resonator (6) and a piezoelectric ceramic (7);

the laser source (1) is configured to output a laser beam;

the laser frequency stabilizer (2) is configured to lock the laser beam to a mode frequency of the optical resonator (6);

the piezoelectric ceramic (7) is configured to adjust a length of the optical resonator (6) to alter the mode frequency of the optical resonator (6) to match energy levels of a target molecular isotopologue;

the signal detector (4) is configured to detect a transmission intensity of the light beam passing the optical resonator (6) to obtain a dual-photon absorption signal; and

the signal analyzer (5) is configured to analyze and process the dual-photon absorption signal to obtain a concentration of the target molecular isotopologue;

wherein the energy levels of the target molecular isotopologue comprise energy levels of dual-photon absorption, and an energy level representing single-photon absorption is between the energy levels of dual-photon absorption; $\frac{1}{2} \Delta E$ characterized in that $\frac{1}{2} \Delta E$ and

~~wherein~~ energy of a photon having the mode frequency matching the energy levels of the target molecular isotopologue is equal to a half of an energy difference between the energy levels of the target molecular isotopologue.

2. The apparatus according to claim 1, wherein the laser beam is a continuous infrared laser beam, and a power of the laser beam is greater than 100mW.

3. The apparatus according to claim 1 or claim 2, wherein a fineness of the optical resonator (6) is higher than 60000.

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4. The apparatus according to any of the preceding claims, wherein fluctuations of temperature of the optical resonator (6) are less than 10mK.

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5. The apparatus according to any of the preceding claims, wherein the sample chamber (3) further comprises:

a temperature controller (10), configured to control temperature of the optical resonator (6).

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6. The apparatus according to any of the preceding claims, wherein:
the signal detector (4) comprises a detector (11) and a signal amplifier (12) which are integrated;

the detector (11) is configured to detect the transmission intensity of the light beam passing the optical resonator (6) to obtain the dual-photon absorption signal;

the signal amplifier (12) is configured to amplify the obtained dual-photon absorption signal.

7. The apparatus according to any of the preceding claims, wherein the signal analyzer (5) is configured to:

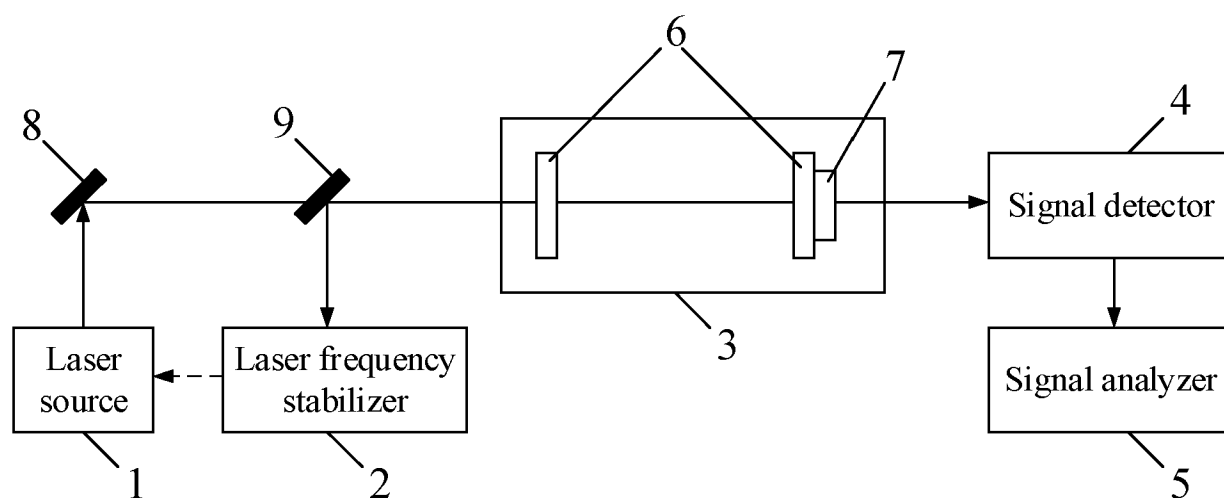
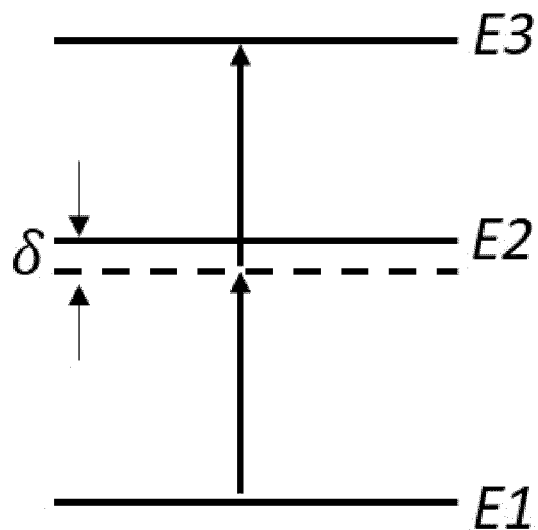
invoke a signal processing program to analyze and process the dual-photon absorption signal to obtain the concentration of the target molecular isotopologue, and

display the concentration of the target molecular isotopologue via an interactive interface.

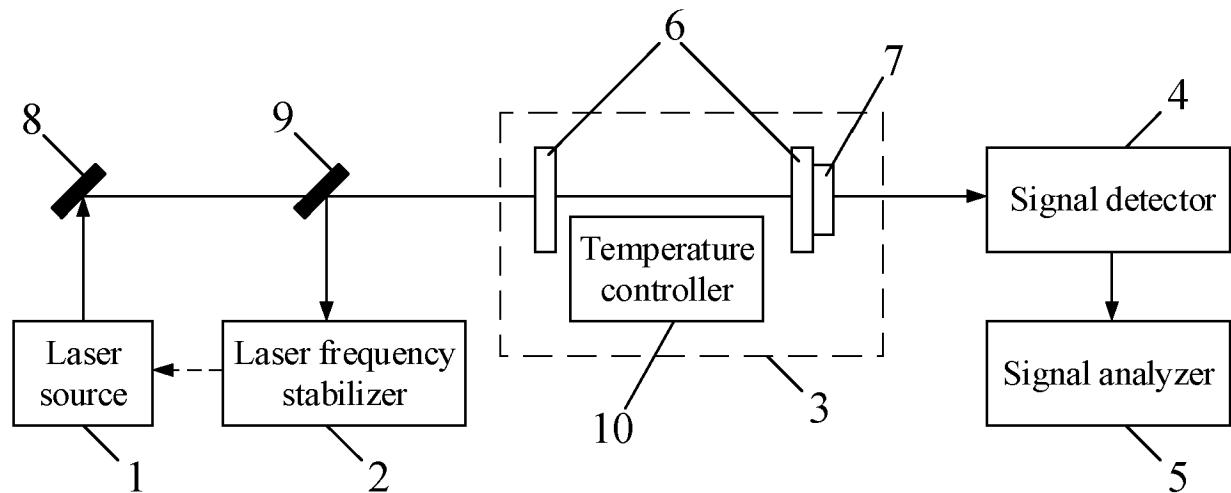
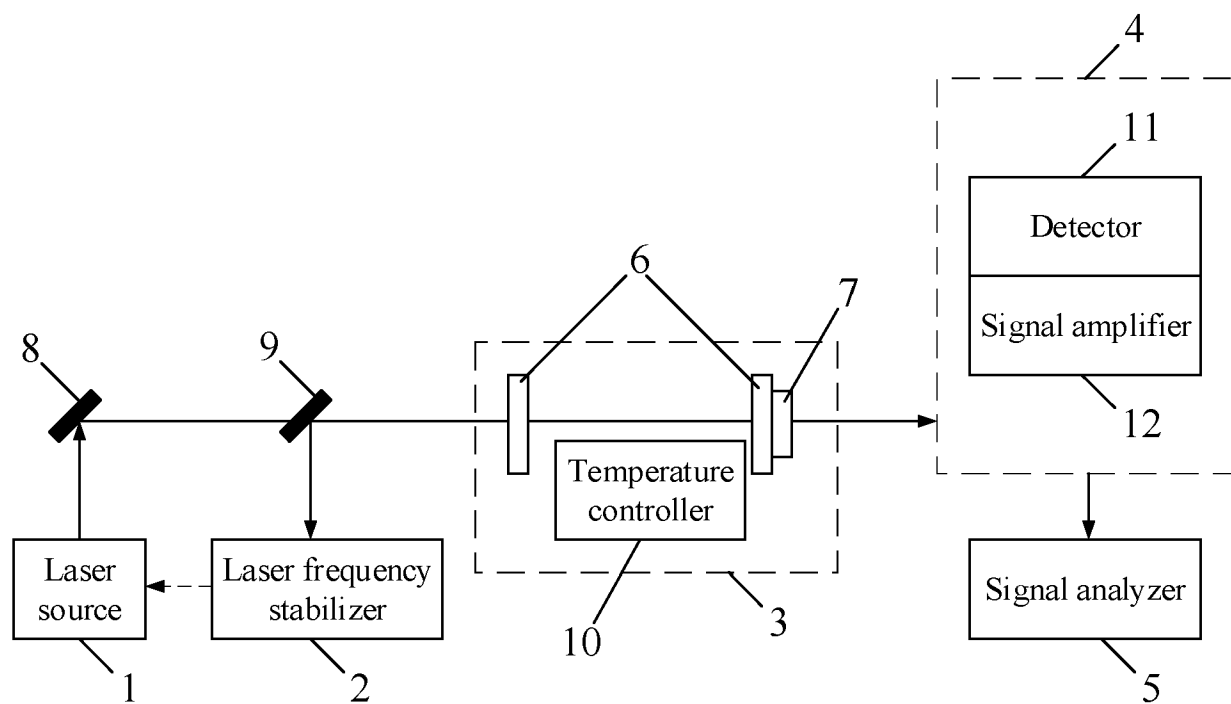
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8. A spectrometer, comprising the apparatus according to any one of claims 1 to 7.

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**Figure 1****Figure 2**

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**Figure 3****Figure 4**

The diagram illustrates the components and signal flow of a laser-based measurement system. The system includes a **Laser source** (1) and a **Laser frequency stabilizer** (2). The laser beam (3) is directed through a series of mirrors (4, 5, 6, 7, 8, 9) and is focused onto a **Temperature controller** (10) which is part of a **Microfluidic device** (3). The device is connected to a **Detector** (11) and a **Signal amplifier** (12). The output of the signal amplifier is processed by a **Signal processing program** (13) and an **Interactive interface** (14).

The diagram illustrates a laser-based spectrometer system. The components are numbered 1 through 9. The system is enclosed in a dashed box labeled "Spectrometer". The components are:

- 1: Laser source
- 2: Laser frequency stabilizer
- 3: A box containing two vertical rectangular components, labeled 6 and 7.
- 4: Signal detector
- 5: Signal analyzer
- 8: A beam splitter or mirror.
- 9: A beam splitter or mirror.

The optical path is as follows:

- The laser source (1) emits a beam that passes through a beam splitter (8).
- The beam then passes through a beam splitter (9).
- The beam enters the box (3) and passes through two vertical rectangular components (6 and 7).
- The beam then enters the signal detector (4).
- The signal detector (4) outputs a signal to the signal analyzer (5).

The laser frequency stabilizer (2) is connected to the laser source (1) and the beam splitter (9).

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