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AUTOIONIZING RESONANCES IN THREE-PHOTON IONIZATION SPECTRUM OF THE YTTERBIUM ATOM

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The three-photon ionization spectrum of ytterbium atoms in the interval 16814.4–18100.0 cm⁻¹ has been studied. In addition to the resonance maxima related to the one- and two-photon excitations of bound states, the maxima originated from the three-photon excitation of autoionizing states are observed. Two new odd autoionizing levels with energies of 51866.1 and 51914.1 cm⁻¹ are discovered.

Keywords: ytterbium, three-photon ionization, autoionizing level.

1. Introduction

The excitation of autoionizing states is of considerable interest from the viewpoint of fundamental issues in the physics of multiphoton processes. The multiphoton resonance with an autoionizing level, which occurs at the final stage of the photon absorption (the k -photon resonance at the k -photon ionization), substantially differs from the well-studied case of the intermediate multiphoton resonance with a discrete bound level (the k_1 -photon resonance at the k -photon ionization, where $k_1 < k$) [1–3]. Unlike the bound levels, the autoionizing ones, which are also atomic discrete levels, lie above the ionization potential. Therefore, as a result of the interelectron configuration interaction, they can spontaneously decay with a high probability into a continuum absorbing no additional photons.

The autoionizing levels play a crucial role in the realization of the two-electron mechanism of two-charge ion formation, which is characterized by a simultaneous detachment of two electrons from a neutral atom. In this case, the ionization process is associated with the excitation of states in the neutral atom, which are strongly perturbed owing to the dynamic Stark effect, and their ionization occurs by means of one-photon transitions in the spectrum of autoionizing states [4].

The role of autoionizing states in multiphoton processes and their behavior in the field of intense laser radiation have been studied theoretically quite thoroughly (see, e.g., works [5–8]), whereas the experi-

mental data are scarce [9–13]. This circumstance is related to the fact that, as a rule, the autoionizing levels weakly, or even in no way, manifest themselves in multiphoton ionization spectra. A possible reason for that may be a substantial broadening of autoionizing levels in the field of intense laser radiation [9]. In addition, the self-manifestation of autoionizing levels in multiphoton ionization spectra is considerably affected by the presence of a quasiresonance between the autoionizing and bound atomic levels [10]. However, the lack of corresponding experimental data does not allow any generalizing conclusions concerning the origin of such a behavior of autoionizing levels to be drawn. Therefore, obtaining new experimental data on the excitation of autoionizing levels at the multiphoton ionization is challenging.

In this work, the spectrum of three-photon ionization of ytterbium atoms is studied in the spectral interval from 16814.4 to 18100 cm⁻¹ containing low-located odd autoionizing levels with energies of 50443.2–54300 cm⁻¹ [14–20], which can be excited by three photons. The data available at present on autoionizing levels from the indicated range were obtained within the photoabsorption method [17, 20], as well as emission [19], photoionization [14], and three-stage resonance-ionization spectroscopies [15, 16, 18]. Their analysis demonstrates that, for a number of levels, the corresponding energies determined with the use of different schemes of their excitation are substantially different. Therefore, the determination of energies for those levels by schemes for their excitation that would be different from those used in works [14–20] is also challenging.

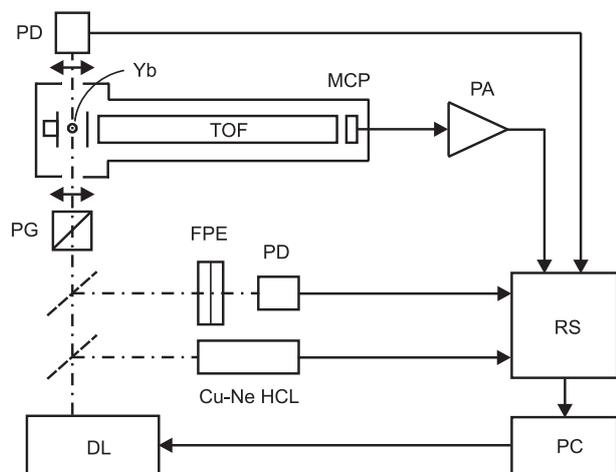


Fig. 1. Schematic diagram of the experimental installation: (DL) dye laser, (Cu-Ne HCL) Cu-Ne hollow-cathode lamp, (FPE) Fabry-Pérot etalon, (PD) photo diode, (PG) polarizer, (TOF) time-of-flight mass spectrometer, (MCP) microchannel detector, (PA) preamplifier, (RS) multichannel registration system, (PC) personal computer

Note also that at present, except for works [11, 21], there are no other data concerning the three-photon ionization of ytterbium atoms in the examined spectral interval. However, in work [21], measurements were carried out at a much higher field strength (about 10^7 V/cm) than in our experiment. At the indicated field strength, the energy level spectrum of the ytterbium atom is considerably perturbed, which manifests itself in a substantial smoothness of the resonance structure observed in work [21]. In particular, in the cited work, unlike this one, no maxima associated with the three-photon excitation of autoionizing levels were revealed. Concerning work [11], the spectral interval $17850\text{--}18180$ cm^{-1} researched there was too narrow.

2. Experiment

The scheme of the experimental installation is shown in Fig. 1. Linearly polarized radiation emitted by a pulsed dye laser FL-2001 (Lambda Physik) with a linewidth of about 0.2 cm^{-2} was focused into a vacuum chamber, where it intersected at the right angle with a beam of ytterbium atoms formed with the use of an effusion source. Ytterbium atoms, which were formed in the region of interaction between the atomic and laser beams, were drawn out with the help of a constant electric field and, after their pass-

ing through a time-of-flight mass spectrometer, were detected using a microchannel detector of the VEU-7 type. The concentration of ytterbium atoms in the interaction region was of an order of 10^{10} cm^{-3} . The residual pressure in the vacuum chamber was about 10^{-5} Pa. Ytterbium atoms became ionized as a result of the absorption of three photons, i.e., the three-photon ionization took place.

The absolute value of laser radiation frequency was determined with an accuracy of not worse than ± 0.3 cm^{-1} from the optogalvanic spectrum of a Cu-Ne hollow-cathode discharge lamp and the transmission spectrum of a Fabry-Pérot interferometer. The energy in each laser pulse was measured using a photo diode with calibrated spectral sensitivity. All signals measured in the course of the experiment were applied to the input of the registration system, transformed there into a digital code, and read out by a computer controlling the whole experimental procedure.

In the experiment, the dependence of the Yb^+ ion yield on the laser radiation frequency, $N^+(\omega)$, was measured in the spectral interval $16814.4\text{--}18100$ cm^{-1} . The lower limit of the interval was selected according to the $IP/3$ -condition, where $IP \approx 50443.1$ cm^{-1} is the first ionization potential of the Yb atom. A value of 18100 cm^{-1} is the upper limit of the generation range by Rhodamine 6G dye. The strength of the laser radiation field in the interaction region was $\varepsilon \approx 10^5$ V/cm. As the frequency ω varied within the interval $16814.4\text{--}18100$ cm^{-1} , the field strength was maintained constant. At such value of ε , the resonance structure associated with the three-photon excitation of autoionizing levels is the most pronounced in the dependence $N^+(\omega)$. In particular, the number of corresponding peaks was maximal in this case. Both the higher and lower values of field strength resulted in a less pronounced manifestation of the resonance structure, in particular, the number of maxima associated with the three-photon excitation of autoionizing levels became smaller. This fact was connected, in the first case, with a considerable broadening of the autoionization peaks and, in the second case, with a reduction of the ion signal ($N^+ \sim \varepsilon^6$).

3. Results of Researches

The measured dependence $N^+(\omega)$ is shown in Fig. 2. The observed resonance structure contains

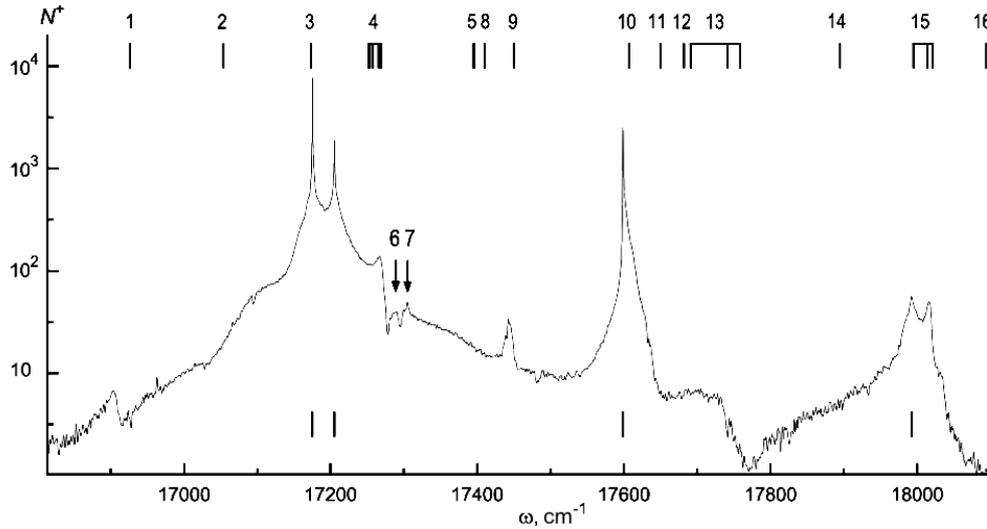


Fig. 2. Spectrum of the three-photon ionization of ytterbium atoms in the interval 16814.4–18100 cm^{-1}

eleven well-resolved maxima with different amplitudes and shapes. The maxima are a manifestation of the one- and two-photon excitations of even bound states and the three-photon excitation of autoionizing states of the ytterbium atoms.

3.1. One- and two-photon excitation of bound states

The data for the maxima corresponding to the one- and two-photon excitations of bound states are quoted in Table 1. In Fig. 2, the frequencies of corresponding one- and two-photon transitions are marked by vertical strokes in the lower part of the figure. In general, the dependence $N^+(\omega)$ reveals two maxima associated with one-photon transitions and two maxima associated with two-photon transitions. The maxima corresponding to two-photon transitions are characterized by the largest amplitudes and the narrowest widths (about 0.4 cm^{-1}). One of them (at $\omega = 17175.3 \text{ cm}^{-1}$) results from the excitation of the outer s -electron ($6s \rightarrow 7s$), whereas another one (at $\omega = 17598.6 \text{ cm}^{-1}$) from the excitation of an electron in the inner f -shell ($4f \rightarrow 6p$) [22]. As one can see from Fig. 2, both maxima are comparable by amplitude. This fact testifies to a high efficiency of the resonance three-photon ionization channel connected with the two-photon excitation of levels in the so-called “complex” spectrum of the ytterbium atoms associated with the excitation of electrons just in the

inner f -shell [23]. Note that both maxima are a result of the excitation of even bound states.

Concerning the maxima generated by one-photon transitions, one of them (at $\omega = 17992.0 \text{ cm}^{-1}$) corresponds to the transition between the ground even level $4f^{14}6s^2 \ ^1S_0$ and the excited odd level $4f^{14}6s6p \ ^3P_1^\circ$, whereas the other (at $\omega = 17205.0 \text{ cm}^{-1}$) is a result of the one-photon transition between two excited levels, the odd $4f^{14}6s6p \ ^3P_1^\circ$ and even $4f^{13}6s^26p \ ^1D_2$ ones [22]. The lower $4f^{14}6s6p \ ^3P_1^\circ$ level becomes populated owing to the hyper-Raman scattering

$$4f^{14}6s^2 \ ^1S_0 + 2\omega - \nu \rightarrow 4f^{14}6s6p \ ^3P_1^\circ$$

($\nu = 16418 \text{ cm}^{-1}$).

Note that the process of Yb^+ ion generation at the frequency $\omega = 17205.0 \text{ cm}^{-1}$ is a four-photon one

Table 1. Identification of maxima in the dependence $N^+(\omega)$ as one- and two-photon transitions

ω, cm^{-1}	Transition
17175.3	$4f^{14}6s^2 \ ^1S_0 + 2\omega \rightarrow 4f^{14}6s7s \ ^1S_0$
17205.0	$4f^{14}6s6p \ ^3P_1^\circ + \omega \rightarrow 4f^{13}6s^26p \ ^1D_2$
17598.6	$4f^{14}6s^2 \ ^1S_0 + 2\omega \rightarrow 4f^{13}6s^26p_{3/2} \ (7/2, 3/2)_2$
17992.0	$4f^{14}6s^2 \ ^1S_0 + \omega \rightarrow 4f^{14}6s6p \ ^3P_1^\circ$

$(2\omega + \omega + \omega):$

$$4f^{14}6s^2\ ^1S_0 + 2\omega - \nu \rightarrow 4f^{14}6s6p\ ^3P_1^\circ + \omega \rightarrow$$

$$\rightarrow 4f^{13}6s^26p\ ^1D_2 + \omega \rightarrow 4f^{14}6s\ ^2S_{1/2} + e.$$

Despite that, the amplitude of the maximum that corresponds to this process is considerable (see Fig. 2). This fact testifies to a high efficiency of the hyper-Raman scattering in this case, which, in our opinion, is induced by the closely located frequencies ($\Delta\omega \approx 29.7\text{ cm}^{-1}$) of the one- ($4f^{14}6s6p\ ^3P_1 \rightarrow 4f^{13}6s^26p\ ^1D_2$, $\omega = 17205.0\text{ cm}^{-1}$) and two-photon ($4f^{14}6s^2\ ^1S_0 \rightarrow 4f^{14}6s7s\ ^1S_0$, $\omega = 17175.3\text{ cm}^{-1}$) transitions.

3.2. Three-photon excitation of autoionizing states

According to the available data on the odd autoionizing levels in the ytterbium atom [14–20], the examined spectral interval $\omega = 16814.4\div 18100\text{ cm}^{-1}$ ($3\omega = 50443.2\div 54300\text{ cm}^{-1}$) includes 10 levels with the total angular momenta $J = 1$ and 3, which, according to the selection rules, can be excited by three photons. In addition, the indicated interval also includes four levels from work [16]. In the cited work, a three-stage excitation scheme for autoionizing states was used,

$$6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1 \rightarrow 6s7s\ ^3S_1 \rightarrow \text{autoionizing states.}$$

This allowed the levels with the total angular momenta $J = 0\div 2$ to be observed. Note that the exact values of J for the considered levels were not determined in work [16]. According to the selection rules, the three-photon excitation of those levels could take place in our experiment only provided that their total angular momentum equals $J = 1$. The data for all odd autoionizing levels in the interval $3\omega = 50443.2\div 54300\text{ cm}^{-1}$ are quoted in Table 2. In Fig. 2, the frequencies corresponding to the three-photon excitation of the known autoionizing levels are marked by vertical strokes in the upper part of the figure.

Note that, at present, electron configurations and terms are not determined for all levels quoted in Table 2. For the majority of them, only the total angular momentum J was found. Moreover, for some levels, the energies determined by different authors,

who used different schemes for the excitation of levels, substantially diverge. In particular, the maximum difference amounts to 49 cm^{-1} for the level $5d6p\ [1/2]_1^\circ$ (No. 4) [15, 20] and to 79 cm^{-1} for the level $4f^{13}5d^26s\ ^5D_1^\circ$ (No. 15) [14, 20], whereas, for the level $5d6p\ [3/2]_1^\circ$ (No. 13), it reaches 203 cm^{-1} [14, 17]. In Fig. 2, the data obtained by different authors for the same levels are connected by horizontal strokes.

Of fourteen known autoionizing levels, only five manifest themselves in the form of pronounced maxima in the measured dependence $N^+(\omega)$ (see Fig. 2). The frequencies of those maxima and the determined energies of autoionizing levels are also quoted in Table 2. A comparison of our values obtained for the energy with the data of other authors shows that, if the divergences discussed above are taken into account, the corresponding values are in rather good agreement with one another.

By their height, all maxima associated with autoionizing states are substantially lower than the maxima corresponding to the two-photon excitation of even bound states, but they are considerably broader. The widest is the maximum at $\omega = 17700.0\text{ cm}^{-1}$ (about 72 cm^{-1}) associated with the excitation of the level $5d6p\ [3/2]_1^\circ$ (No. 13). The widths of all other maxima do not exceed 20 cm^{-1} . Note that the width of the level $5d6p\ [3/2]_1^\circ$ determined by the authors of works [14] and [17] amounts to 542 and 125 cm^{-1} , respectively, whereas the widths of other discussed levels are much narrower falling within the interval $14\text{--}58\text{ cm}^{-1}$ [14, 15, 17].

From Fig. 2, one can see that the amplitude of the maximum at $\omega = 18016.4\text{ cm}^{-1}$ associated with three-photon excitation of the autoionizing level $4f^{13}5d^26s\ ^5D_1^\circ$ (No. 15) is comparable with the amplitude of the maximum at $\omega = 17992.0\text{ cm}^{-1}$ corresponding to the one-photon resonance at the three-photon ionization. In our opinion, this is a result of the small difference ($\Delta\omega \approx 24.4\text{ cm}^{-1}$) between the frequencies of the one-photon transition $4f^{14}6s^2\ ^1S_0 \rightarrow 4f^{14}6s6p\ ^3P_1$ and the three-photon transitions $4f^{14}6s^2\ ^1S_0 \rightarrow 4f^{13}5d^26s\ ^5D_1$.

The analysis of the available data on the spectrum of bound and autoionizing levels of the ytterbium atoms [14–20, 22, 24, 25] testifies that two maxima observed in the dependence $N^+(\omega)$ at frequencies of 17288.0 and 17304.7 cm^{-1} are not a result of the excitation of known levels. In Fig. 2, they are marked

Table 2. Data for odd autoionizing levels of the ytterbium atoms in the interval 50443.2–54300 cm⁻¹

No.	Levels	$E, \text{cm}^{-1} / (\omega, \text{cm}^{-1})$							
		Our data	[14]	[15]	[16]	[17]	[18]	[19]	[20]
1	$4f^{13}5d^2ns[k]_1^\circ$	50711.4 (16903.8)				50779.5 (16926.5)			
2	$J = 1$				51162.6 (17054.2)				
3	$J = 1$			51520.1 (17173.4)					
4	$5d6p[1/2]_1^\circ$	51801.0 (17267.0)	51771 (17257.0)	51805.2 (17268.4)		51797 (17265.7)		51760 (17253.3)	51756 (17252.0)
5	$J = 3$			52187.2 (17395.7)			52186 (17395.3)		
6*	$J = 1, 3$	51866.1 (17288.7)							
7*	$J = 1, 3$	51914.1 (17304.7)							
8	$4f^{13}6s^26d[3/2]_1^\circ$					52230 (17410.0)			
9	$J = 3$	52327.2 (17442.4)		52349.9 (17450.0)					
10	$J = 0-2$				52821.7 (17607.2)				
11	$J = 0-2$				52951.3 (17650.4)				
12	$J = 0-2$				53047.0 (17682.3)				
13	$5d6p[3/2]_1^\circ$	53100.0 (17700.0)	53073 (17691.0)			53276 (17758.7)		53226 (17742.0)	53225 (17741.7)
14	$J = 0-2$				53684.2 (17894.7)				
15	$4f^{13}5d^26s^5D_1^\circ$	54049.2 (18016.4)	53985 (17995.0)			54042 (18014.0)		54042 (18014.0)	54064 (18021.3)
16	$4f^{13}5d^26s^3S_1^\circ$		54281 (18093.7)						

by arrows. With regard for the magnitude and the shape of those maxima, we may assert that they are associated with the three-photon excitation of autoionizing levels, which are absent in the literature at present. The energies of those levels equal 51866.1 and 51914.1 cm⁻¹, respectively, and the total angular momenta may be $J = 1$ or 3 in accordance with the selection rules for three-photon transitions. In Table 2, the data for those levels (No. 6 and 7) are marked with an asterisk (*) and emphasized in bold.

Nine autoionizing levels, the data for which are contained in Table 2 (No. 2, 3, 5, 8, 10–12, 14, and 16), do not manifest themselves as well-distinguished

maxima in the dependence $N^+(\omega)$ (see Fig. 2). A low probability of their three-photon excitation may be a possible reason for that. In addition, as was mentioned above, four levels from work [16] (No. 10–12 and 14) can be excited by three photons only provided that their total angular momentum equals $J = 1$. Therefore, the reason for why the maxima associated with their excitation are absent in our case may be the fact that the total angular momentum for each of those levels equals $J = 0$ or 2.

However, it should be noted that it is difficult to assert unambiguously in some cases that the autoionizing level is not excited or that the excitation does take

place but does not manifest itself as a pronounced maximum in the dependence $N^+(\omega)$. First of all, this concerns levels Nos. 2, 3, 10–12, and 14 (Table 2).

For example, the nonmonotonic behavior of the dependence $N^+(\omega)$ in the frequency interval 17030–17130 cm^{-1} (see Fig. 2) can be a manifestation of three-photon excitations of level No. 2. According to the spectra presented in work [16], the width of this level is approximately equal to 20–30 cm^{-1} , which agrees rather well with a peculiarity observed in this interval.

From Fig. 2, one can easily see that, a frequency of 17173.4 cm^{-1} , which corresponds to the three-photon excitation of autoionizing level No. 3, practically coincides with the frequency of the two-photon excitation of the even bound level $4f^{14}6s7s\ ^1S_0$ ($\omega = 17175.3\ \text{cm}^{-1}$). For this reason, a weak maximum that corresponds to the autoionizing level excitation can simply not manifest itself against the much more intense two-photon maximum.

A similar situation takes place in the case of levels Nos. 10 and 11. The frequencies of their three-photon excitation, 17607.2 and 17650.4 cm^{-1} , respectively, are located in a vicinity of another intense maximum at $\omega = 17598.6\ \text{cm}^{-1}$ associated with the two-photon transition $4f^{14}6s^2\ ^1S_0 \rightarrow 4f^{13}6s^26p_{3/2}\ (7/2, 3/2)_2$. The asymmetry of this maximum on its high-frequency side (see Fig. 2) can be explained as a contribution of the indicated autoionizing levels, which, at the same time, do not manifest themselves in the form of more or less pronounced peaks against the background produced by the maximum. According to the spectra presented in work [16], the widths of levels Nos. 10 and 11 do not exceed 2 cm^{-1} , which agrees well with the observed asymmetry of the maximum concerned.

Level No. 12 is also narrow, with a width of 2 cm^{-1} [16]. Moreover, its energy does not differ strongly from the energy of level No. 13 (see Table 2). Therefore, in view of the total error of different experiments, which is observed practically for all data quoted in Table 2, we may suppose that they are the same level. However, the considerably different level widths, which were observed in works [14] (about 542 cm^{-1}) and [16] (about 2 cm^{-1}) allows us to assert that these are two different levels. A small difference between the three-photon excitation frequencies of levels Nos. 12 ($\omega = 17682.3\ \text{cm}^{-1}$) and 13 ($\omega = 17691.0\ \text{cm}^{-1}$) together with the consid-

erable width of level No. 13 can result in that the corresponding maxima are not resolved, but manifest themselves as a single wide maximum (see Fig. 2). In this case, it is impossible to establish unambiguously whether level No. 12 was excited or not.

From Fig. 2, one can see that the maximum at $\omega = 17992.0\ \text{cm}^{-1}$, which corresponds to the one-photon transition $4f^{14}6s^2\ ^1S_0 \rightarrow 4f^{14}6s6p\ ^3P_1$, is asymmetric on its low-frequency side. This section includes the three-photon excitation frequency of level No. 14 ($\omega = 17894.7\ \text{cm}^{-1}$). However, according to the spectra presented in work [16], the width of level No. 14 does not exceed 2 cm^{-1} . This fact does not allow us to explain the substantial asymmetry of the considered maximum within the interval about 200 cm^{-1} in width exclusively by the three-photon excitation of narrow autoionizing level No. 14. The observed asymmetry can be a result of the three-photon excitation of a wider autoionizing level or even several levels, the data on which are absent in the literature at present. However, it is extremely difficult to determine the energy of that level (or the energies of those levels) proceeding from the behavior of dependence $N^+(\omega)$ in the interval 17776–17970 cm^{-1} . In this situation, it is impossible to judge unambiguously whether level No. 14 is excited or not.

4. Conclusions

The spectrum of the three-photon ionization of ytterbium atoms in the interval 16814.4–18100 cm^{-1} has been studied. The resonance structure of the dependence $N^+(\omega)$ results from the one- and two-photon excitations of bound states and the three-photon excitation of autoionizing states. The maxima associated with the excitation of autoionizing states are several orders of magnitude lower by amplitude than the maxima corresponding to the two-photon excitation of bound states, but considerably exceed them by width. Two unknown odd autoionizing levels were revealed. Their energies equal 51866.1 and 51914.1 cm^{-1} , and the probable values of total angular momentum are $J = 1$ and 3.

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АВТОІОНІЗАЦІЙНІ РЕЗОНАНСИ
В СПЕКТРІ ТРИФОТОННОЇ ІОНІЗАЦІЇ
АТОМА ІТЕРБІЮ

Резюме

Досліджено спектр трифотонної іонізації атома ітербію в діапазоні 16814,4–18100,0 см⁻¹. Поряд з резонансними максимумами, зумовленими одно- та двофотонним збудженням зв'язаних станів, спостерігалися максимуми, що є результатом трифотонного збудження автоіонізаційних станів. Виявлено два нових непарних автоіонізаційних рівня з енергіями 51866,1 см⁻¹ та 51914,1 см⁻¹.