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RESONANCE PHENOMENA IN THE MULTIPHOTON IONIZATION OF EXCITED ATOMS

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RESONANCE PHENOMENA IN THE MULTIPHOTON IONIZATION OF EXCITED ATOMS

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Invited lecture presented at the University of Windsor and at the University of Waterloo /Canada/

ABSTRACT

The experimentally measured power, k, in the power function dependence of the multiphoton ionization probability $W \sim \alpha I^k$ has been found in this institute laboratory to be higher than the theoretical value of the number of photons, k_o, absorbed in the ionization process, contrary to the result $k < k_o$ of previous measurements. The variation of the power in different experimental circumstances is explained by multiphoton resonances with the atomic levels. The general dependence of the multiphoton ionization probability on the light intensity, measured over a wide range, deviates strongly from the power law dependence and can be used to determine the Stark shift of the atomic states involved.

ÖSSZEFOGLALÁS

A többfotonos ionizáció valószinüsége $(W \sim \alpha I^k)$ k kitevőjére kisérletileg először adódott az elméleti k_o értéknél nagyobb érték, mig az irodalomban k_o-nál kisebb értéket több izben is kimutattak. k_o értéke a többfotonos ionizáció folyamatában abszorbeált kvantumok számával egyezik meg. A hatványkitevő különböző paraméterektől való függését a többfotonos rezonancia jelenségével magyarázzuk meg.

Mértük a többfotonos ionizáció valószinüségének a fényintenzitástól való függését nagy fényintenzitás-tartományban és ez a függés a hatványfüggéstől erősen eltér. A mért görbéket az atomi nivók fénytérben való energiaértékének meghatározására használjuk fel.

РЕЗЮМЕ

Экспериментально измеренное значение k показателя степени в функциональной зависимости $(W \sim \alpha I^k)$ вероятности многофотонной ионизации от интенсивности /I/, оказалось впервые больше теоретически рассчитанного числа фотонов k_o, поглощаемых в процессе ионизации, в отличие от имеющихся в литературе экспериментальных результатов, где $k < k_o$.

Различные значения показателя k, получающиеся при различных условиях эксперимента, объясняются наличием многофотонных резонансов взаимодействия с соответствующими атомными уровнями. В широком интервале интенсивностей измерялись зависимости вероятности ионизации, отличающиеся от степенной функции. Измеренные зависимости использовались для определения штарковских сдвигов атомных уровней.

1. THEORY AND HISTORY

Since the publication of the Maria Göppert's work [1] early in 1929, it has been theoretically well established that atoms can be excited, and indeed ionized, even when their ionization potential /IP/ is greater than the energy of the irradiating light quanta ($\hbar\omega$). In such a case the atom must evidently simultaneously absorb k_o quanta in order to satisfy the conservation of the energy

$$(k_o - 1)\,\hbar\omega \;<\; IP \;<\; k_o\,\hbar\omega \qquad /1/$$

For the observation of such a process, however, we had to wait till the invention of the laser, because the probability of its taking place is very small and therefore fairly high light intensities /I/ are needed. At suitable high light intensities the structure too of the atoms is distorted, and by measuring the characteristics of the multiphoton ionization we can determine the influence not only of the field-free structure of the atom on the multiphoton ionization but also of its distortion.

The probability W of a k_o photon ionization is given by a k_o-order approximation of the perturbation theory in the approximation of electric dipol /P = er E/ interaction

$$W = \alpha\, I^{k_o} \qquad /2/$$

where the "cross-section" of the ionization

$$\alpha \sim \left| \Sigma \frac{\langle f|r|\ell\rangle\langle\ell|r|m\rangle \ldots \langle n|r|i\rangle}{\left[E_\ell - E_o - (k_o-1)\hbar\omega + i\gamma_\ell\right]\cdots\left[E_n - E_o - \hbar\omega + i\gamma_n\right]} \right|^2 \qquad /3/$$

is constant. E is the electric field strength of the light wave, and r^* is the coordinate of the electron.

According to these two expressions /2,3/, the logarithm of the ionization probability depends linearly on the logarithm of the light intensity, and the slope of the resulting line is the number of quanta (k_o) absorbed in the ionization process. But already in the first experimental observations of the multiphoton ionization of rare gases [2] which were made with the experimental setup that can be seen in Fig.1 [3], it turned out that the slope of this line - or in other words the apparent number of quanta absorbed in the ionization process - is always smaller than the theoretical value, k_o /see Table I/.

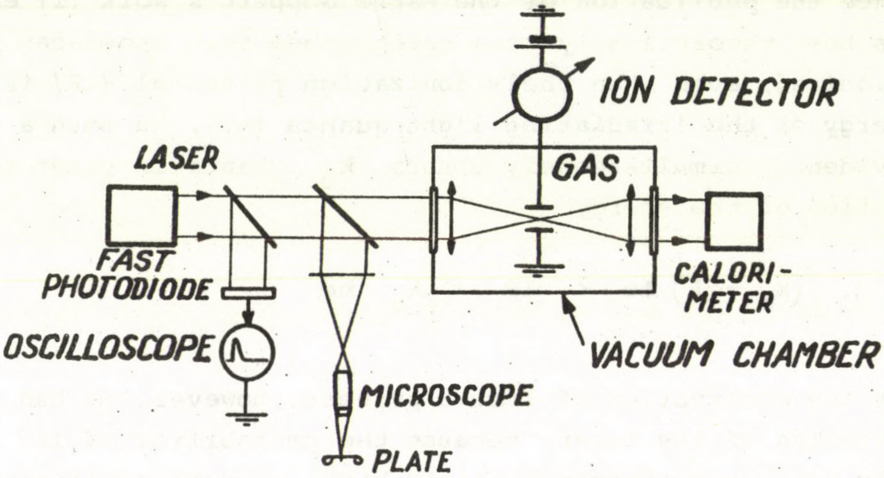


Fig.1

Experimental set-up for measuring the multiphoton ionization of rare gases

In an attempt to explain this effect it was supposed that, in the interaction of atoms having a large ionization potential with the high-intensity light field needed for their ionization, atomic levels near to the ionization threshold become smeared [4] and consequently the effective ionization potential of the atoms decreases [5].

Another cause of observing smaller than the theoretical value of the factor k may be the shift of the atomic level which is in $k < k_o$ photon resonance, i.e. the energy of k quanta coincides with the energy of the level [6].

To verify the correctness of these suppositions first the multiphoton ionization of atoms having smaller ionization potential than the rare gases had to be investigated without multiphoton resonance.

Such atoms are the alkali metals and excited rare gases. The results, which were obtained for the ionization of Na atoms, seemed to justify the assumptions [7]. The dependence of the multiphoton ionization probability of Na atoms agreed with the theoretically expected probability; that is, the measured slope of the line (k) in the logW - logI plot was the same as theoretical value (k_o).

To investigate the influence of the multiphoton resonance on the multiphoton ionization probability the four-photon ionization of the K atom [8] was measured. The energy of three quanta nearly coincides with the energy of the 4f atomic states /three-photon resonance/ /Fig.2/. The slope of the line in the logW - logI plot was measured by tuning the frequency of the laser radiation.

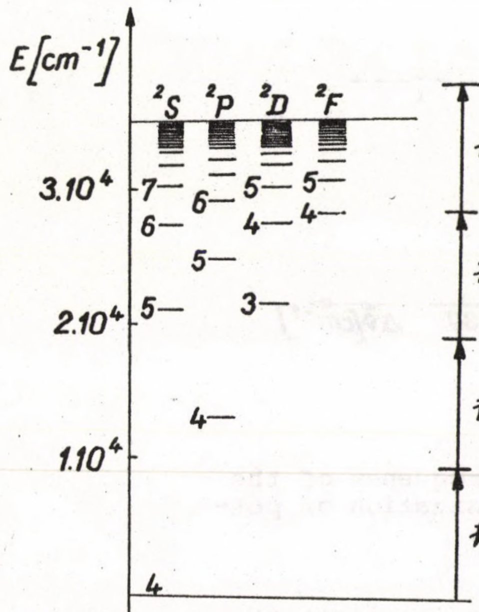


Fig. 2

Multiphoton ionization of potassium atoms: term scheme with the energies of Nd glass laser radiation quanta. The energy of three photons coincides with the energy of the 4^2F state of the atoms

It is easy to deduce from this that in the case of $k < k_o$ photon resonance one of the resonance denominators of expression /3/ approaches zero, and also that the dependence of the energy and width of the resonance level on the light intensity has to be taken into account as a higher than k_o order approximation of the perturbation theory. The "cross section" consequently is a function of the light intensity:

$$\alpha = \alpha(I) \qquad /4/$$

Where

$$E_\ell = E_{\ell o} + \Delta E_\ell(I)$$
$$\gamma_\ell = \gamma_{\ell o} + \Delta\gamma_\ell(I) \qquad /5/$$

$E_{\ell o}$ and $\gamma_{\ell o}$ are the field-free energy and width of the level (ℓ), while $\Delta E_\ell(I)$ and $\Delta\gamma_\ell(I)$ are light-intensity-dependent correction terms.

If this expression of the k_o-photon ionization probability is accepted, the measured curve - being a linear approximation - may be only the tangent of this function in the logW - logI plot. The first derivative of the function /i.e. the slope of the tangent/ was measured in the experiment to be

$$k_{meas} = k = \left(\frac{\partial \log W(I, \Delta\nu_\ell)}{\partial \log I} \right)_{I=const} \qquad /6/$$

$\Delta\nu_\ell = E_{\ell o} - E_{oo} - kh\omega$ is the field-free detuning of level ℓ.

From the theoretical dependence of $\Delta k = k_o - k$ on the field-free detuning presented in Fig. 4 [9] it is apparent that the experimental curve /Fig. 3/ resembles the theoretical curve for $\Delta\gamma > \Delta E$ /curve ② in Fig. 4/.

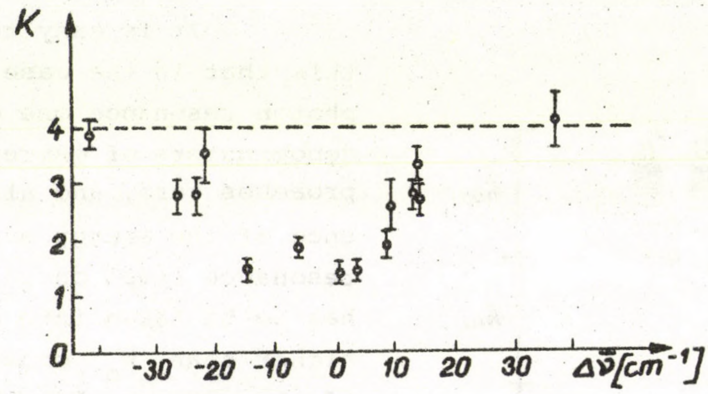


Fig. 3

Dependence of the factor k on the frequency of the laser radiation in the four-photon ionization of potassium atoms |8|

It should be noted that we do not take into account one possible cause of the decrease of the measured slope (k), namely, the saturation due the change of population (N_o) of the initial level of the process, because this is only a trivial experimental difficulty [10]. In point of fact in the experiment only the number of ions (Ni) created by laser pulse is observed, which is proportional to the probability of the ionization

$$N_i = WN_o\tau^{(k)}$$

only in the case when the effectivity of the process /i.e. the saturation parameter/ $W\tau^{(k)} \ll 1$, where $\tau^{(k)}$ is the effective k-photon time duration of the laser pulse.

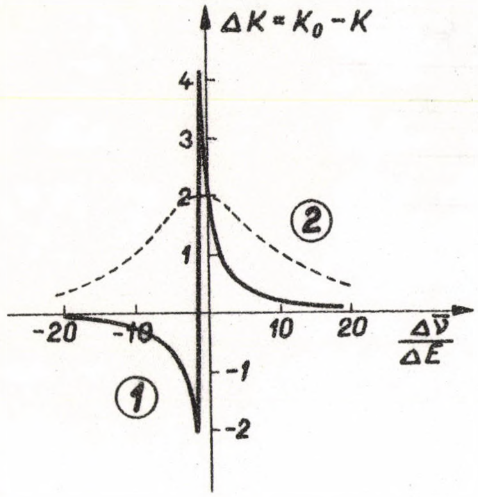


Fig. 4

Theoretical dependence of the factor k on the frequency of the laser radiation ($\Delta\bar{\nu}\sim\bar{\nu}$) at different ratios of the Stark shift /ΔE/ to the level broadening /Δγ/ [8]

2. THE OBJECT OF OUR OWN WORK

With the aim of revealing distortion of atomic structure in the range of the smearing of levels and establishing the role of multiphoton resonance in the multiphoton ionization process in general we have investigated the multiphoton ionization of excited atoms. There was a hope that in this case it would be possible to select the right values of the required ionization potential more easily than in ionization from the ground state.

3. THREE-PHOTON IONIZATION OF METASTABLE HE ATOMS

We first investigated the three-photon ionization of metastable He atoms using the radiation of a ruby laser [11].

The term scheme of He is drawn in Fig. 5. Clearly, there is a quasi-resonance with the 6^1S state of the atom in the case of ionization from the singlet metastable state. As such a resonance is absent in the three-photon ionization of the triplet metastable state, it could be expected that the ionization of the singlet metastable is more probable [12].

The experimental set-up is illustrated in Fig.6 . The laser was Q-switched by a rotating prism and the giant pulse synchronised with the other parts of the measuring set-up /such as the oscilloscope/ by means of a specially constructed electronic synchronizing device [13]. The parameters of the laser pulse, including its energy, the spatial distribution of the light intensity in the focal plane of a 400 mm focal length lens, and the time distribution, were measured in the usual manner [14]. The He atoms were excited by a mild /I = 3mA/ gas discharge in an 11 mm

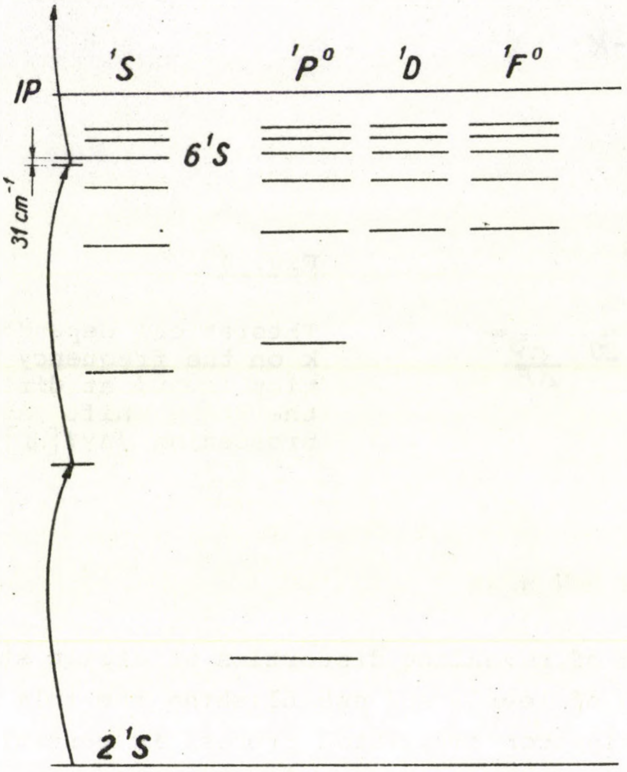


Fig. 5

Term scheme of singlet He atoms with the energies of the ruby laser radiation quanta

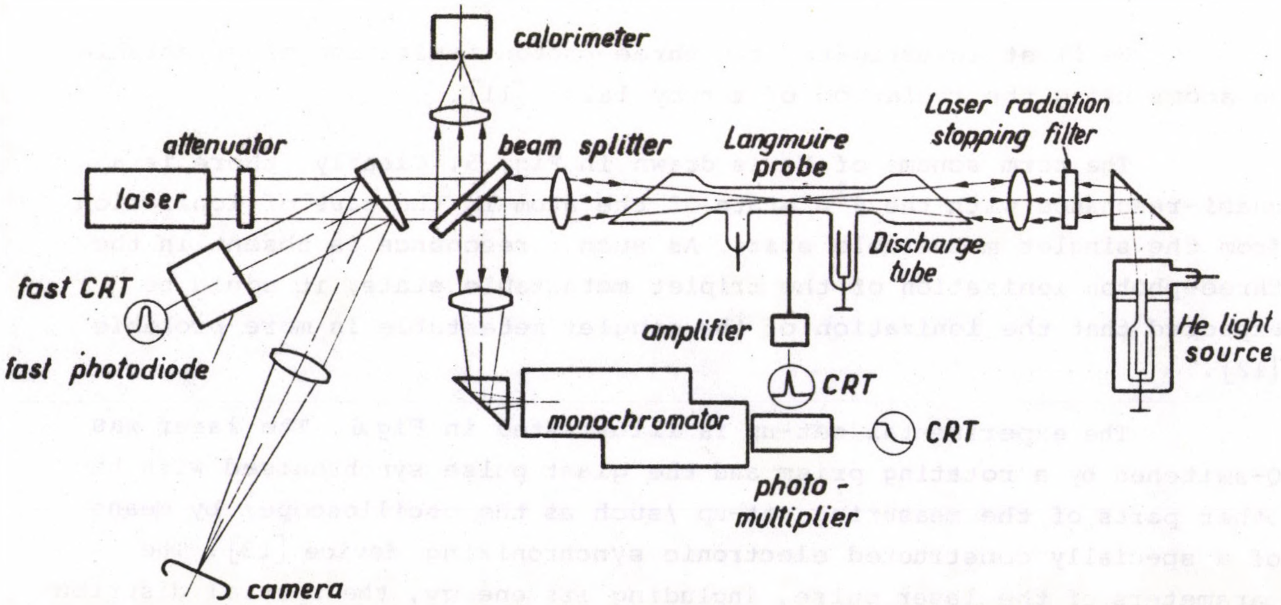


Fig. 6

Experimental set-up for the simultaneous measurement of multiphoton ionization from metastable state of He atoms in the afterglow and of the density and lifetime of the metastables diam /2R/ and about 100 mm long glass tube. The distribution of the metastable density in the cross section of the tube was supposed to be the normal mode of the diffusion $J_o\left(2.4\,\frac{\rho}{R}\right)$. The ions were detected with a Langmuire probe located 1 mm from the axis of the tube [15]. The pressure of the helium gas was about 1.5 mm Hg, and the density of ions during glow discharge $\sim 10^{11}$ cm^{-3}. The density of the metastables N_m was measured to be $\sim 10^{11}$ cm^{-3} by absorption of the line originating from the transition to the metastable state concerned.

The laser light pulse, after being focused into the middle of glass tube by the lens interacts there with the metastables in the afterglow period, at a time Δt after quenching the discharge, and creates excess ions with a distribution [16]

$$f(r,0) = N_m\,(\underline{r})\,\alpha I_o^{k_o}\,g_1^{k_o}\,(\underline{r}) \int g_2\,(t)\,dt \qquad /7/$$

After the laser pulse, the excess ions diffuse and consequently a pulse appears on the probe with an amplitude (V) proportional to the maximum density of excess ions at the probe:

$$V \sim \left(\sum_{i,k} c_{ik}\,J_o\left(\eta_i\,\frac{\rho}{R}\right)\sin\frac{2\pi kz}{L}\,e^{-\frac{t}{\tau_{ik}}}\right)_{max} = B\alpha I^{k_o}\,\tau^{(k_o)} \qquad /8/$$

$$c_{i\ell} = \alpha I^{k_o}\,\tau^{(k_o)} \int N_m(\underline{r})\,J_o\left(\eta_i\,\frac{\rho}{R}\right) g_1^{\,k_o}(\underline{r})\,\sin\frac{2\pi\ell}{L}\,z\cdot d\underline{r} \qquad /9/$$

$$\tau^{(k_o)} = \int g_2^{k_o}(t)\,dt \qquad /10/$$

Here I_o is the peak light intensity, while $\tau_{i\ell}$ is the decay time of the diffusion mode i,ℓ; $g_1(\underline{r})$, $g_2(t)$ are the spatial and temporal intensity distribution

$$\tau_{i\ell} = \frac{D}{p\Lambda^2} \qquad \frac{1}{\Lambda^2} = \left(\eta_i\,\frac{1}{R^2} + \ell\,\frac{1}{L^2}\right)^{1/2} \qquad /11/$$

The theoretically calculated time dependence of the probe pulse and its variation with the position of the probe are plotted in Fig. 7 assuming the model initial distribution of the ion density indicated in the figure [16].

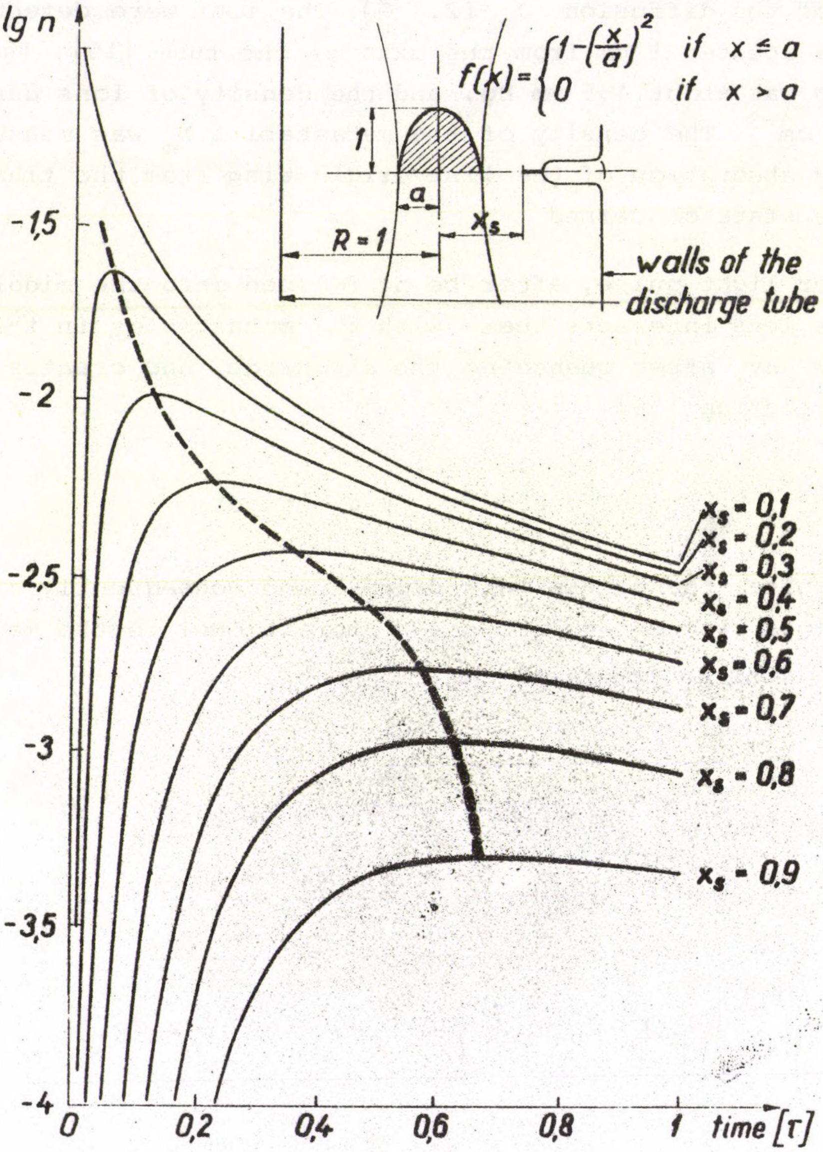


Fig. 7

Dependence of the amplitude /dotted line/ and form of the pulse /full line/ of the Langmuire probe on its location and time, assuming the initial ion density distribution indicated at the top of the figure. The amplitude of the pulse is proportional to the density of ions /n/ at the position of the probe. The time unit is the decay time of the normal mode of the diffusion

The pulse shape photographed on the screen of the cathode ray tube can be seen in Fig.10 on background of decaying ion density of the after-glow plasma [11]. Later in the measurement, the signal was separated from background by a specially constructed electronic detecting device [15]

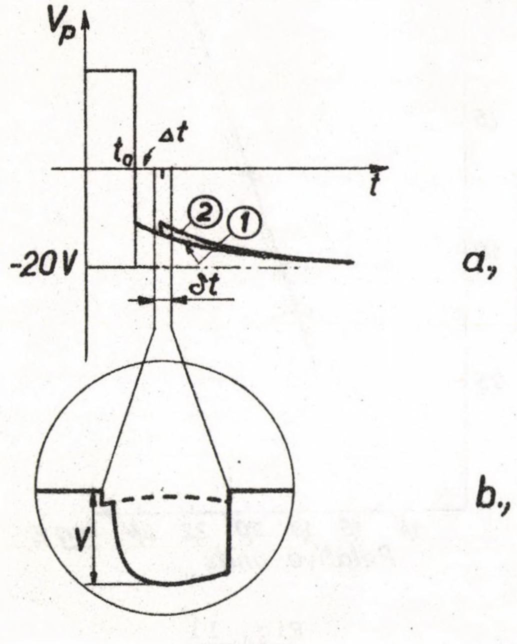
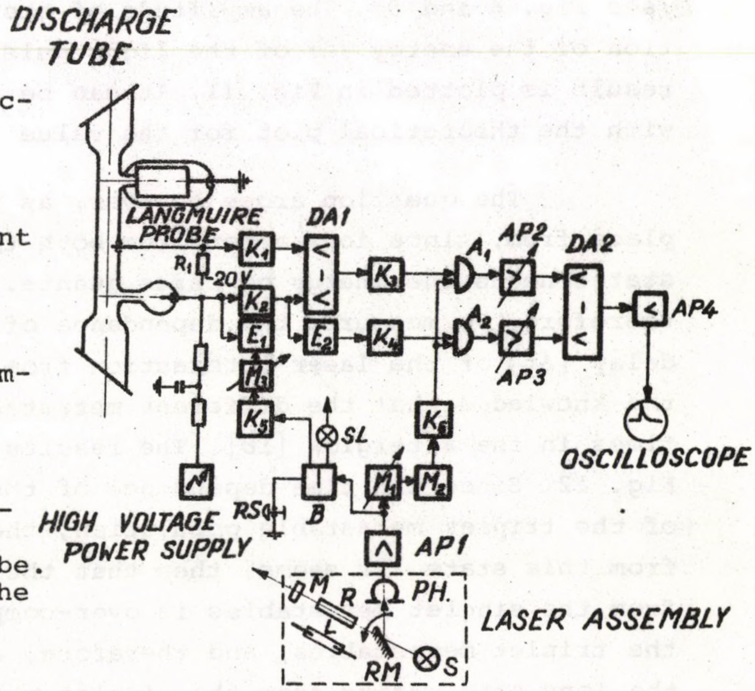


Fig. 8

Multiphoton signal /2/ on the background of the ion signal of the decaying plasma /1/ in the circuit of the Langmuire probe. The step in the potential of the probe /V_p/ is caused by the switching off of the discharge and the consequent drop in plasma potential at the time point t_o. The declining potential does not reach the bias of the probe /-20V/ because the ion signal of the afterglow decays exponentially /1/. Interaction of the laser radiation with the decaying plasma /i.e. with the metastable atoms/ takes place at a time /Δt/ after the discharge has been switched off and the corresponding signal has appeared on the background of the decaying ion signal /2/. An electrical measuring set-up /Fig.9/ selects the time interval/δt/ in which the leading edge of the multiphoton signal occurs, and compensates the background of the decaying plasma. The resulting pulse shape of the multiphoton signal is given by Fig.8/b; the amplitude of the pulse /V/ is proportional to the maximum ion density at the focal point.

Fig. 9

Set-up for measurement of multiphoton ionization of metastable He atoms in afterglow discharge. APi, Ki and Mi are amplifiers, cathode followers and multivibrators, respectively; B is a bistable multivibrator; RS and SL are a ready switch and lamp; R, L, M and S are the laser rod, a flash lamp, the laser exit mirror and an incandescent light source. Switching off of the discharge is initiated by the PH synchronising photodetector/AP1, B, K5, M3, E1/ of a laser Q-switched with a rotating prism /RM/, the timing of switch-off being determined by multivibrator M3. The differential amplifier DA1 compensates the drop in potential of the probe following switch-off /see Fig.8/ using the simultaneous drop in the anode potential of the discharge tube. L2 forms from the voltage drop on the anode of the discharge tube an exponentially decaying pulse having the same shape as the ion signal of the decaying plasma and using this signal differential amplifier DA2 compensates for the ion background.



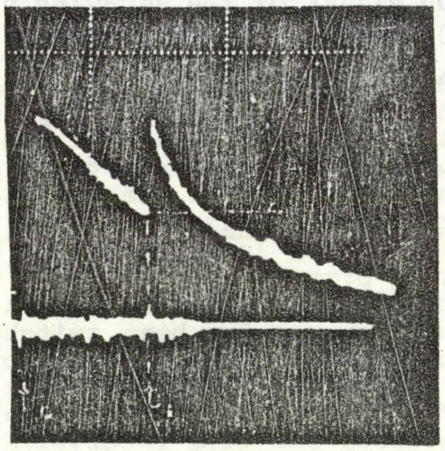


Fig. 10

The multiphoton signal superimposed on the background of the ion signal of the decaying plasma, as it appears on the oscilloscope screen

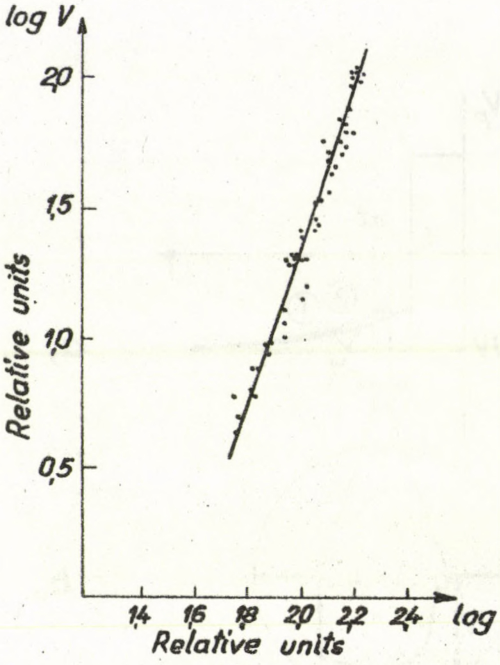


Fig. 11

Dependence of the amplitude of the multiphoton ionization signal of the Laingmuire probe on the energy of the laser pulse /ε/

/see Fig. 8 and 9/. The amplitude of the probe pulse was measured as a function of the energy /ε/ of the light pulse /i.e. the peak intensity/ and the result is plotted in Fig. 11. It can be seen that the slope of the line agrees with the theoretical plot for the value $k_o = 3$ [17].

The question arose however, as to what state the ionization takes place from, since ionization from both the singlet and triplet metastable states needs the energy of three quanta. To distinguish between two processes, therefore, we measured the dependence of the probe pulse amplitude on the delay (Δt) of the laser interaction from the quenching of the discharge, in the knowledge that the different metastable atoms have different mean lifetimes in the afterglow [18]. The results of the measurement can be seen in Fig. 12. Since the time dependence of the multiphoton signal agrees with that of the triplet metastable population, the ionization must obviously take place from this state. It seems, then that the higher probability of ionization from the singlet metastables is over-compensated by the greater density of the triplet metastables, and therefore, at large values of the delay Δt, the ions originating from the singlet metastable states can not be observed.

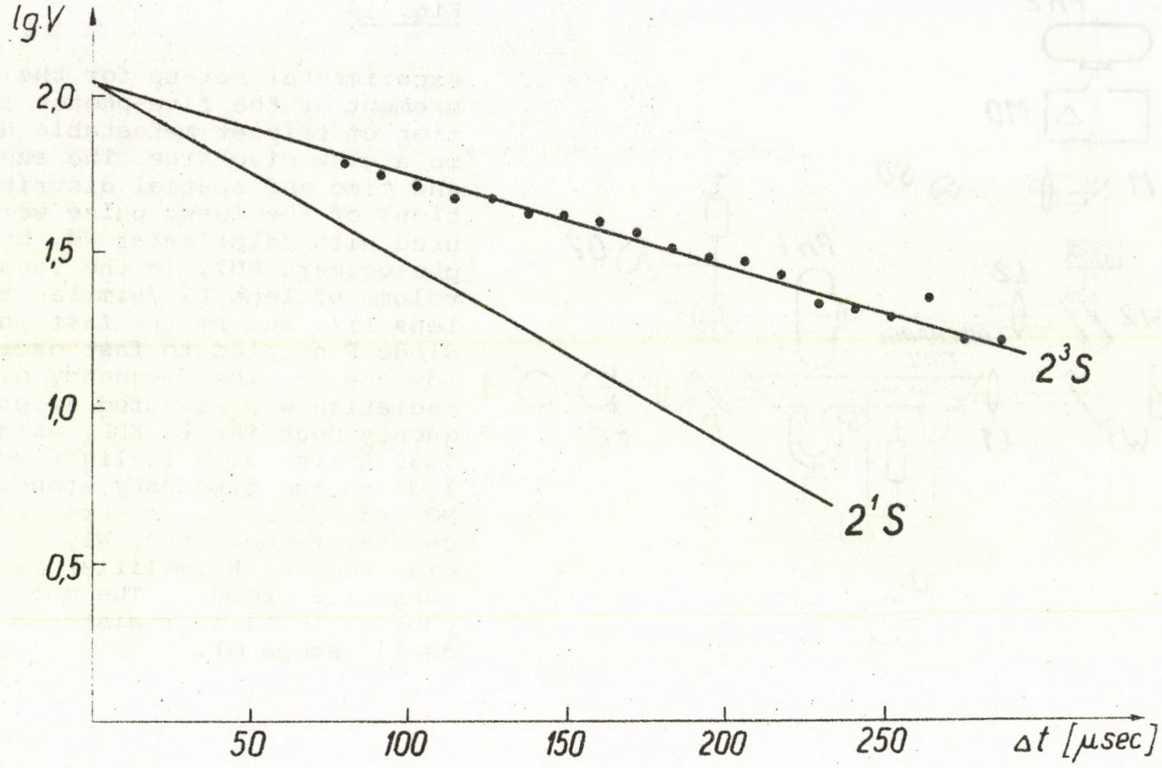


Fig. 12

Dependence of the multiphoton ionization signal on the time delay /Δt/ of the laser pulse after the discharge is switched off

4. FIVE-PHOTON IONIZATION OF TRIPLET METASTABLE HE ATOMS

In continuing this work we studied the dependence of the ionization probability on the light intensity using the radiation of a Q-switched Nd^+ glass laser in a mild gas discharge. The experimental apparatus, which is sketched in Fig. 13, did not differ essentially from the previous set-up [19].

The energy of the ionizing quanta in the term scheme of the atom can be seen in Fig. 14. In this case resonance would plainly be expected in the triplet system very near to the ionization threshold. The position of the fourth quanta in relation to the atomic levels is also plotted in enlarged scale. Ionization from the singlet metastable state needs four, and that from the triplet state five, quanta. Now it is easy to differentiate one process from the another. The result of the measurement is seen in Fig. 15. The slope of the line in the logW - logI plot is $k = k_o = 5$. This agrees with the theoretical value of five-photon ionization from the triplet metastable states, which is the process, to be expected from the resonances in the triplet system [20].

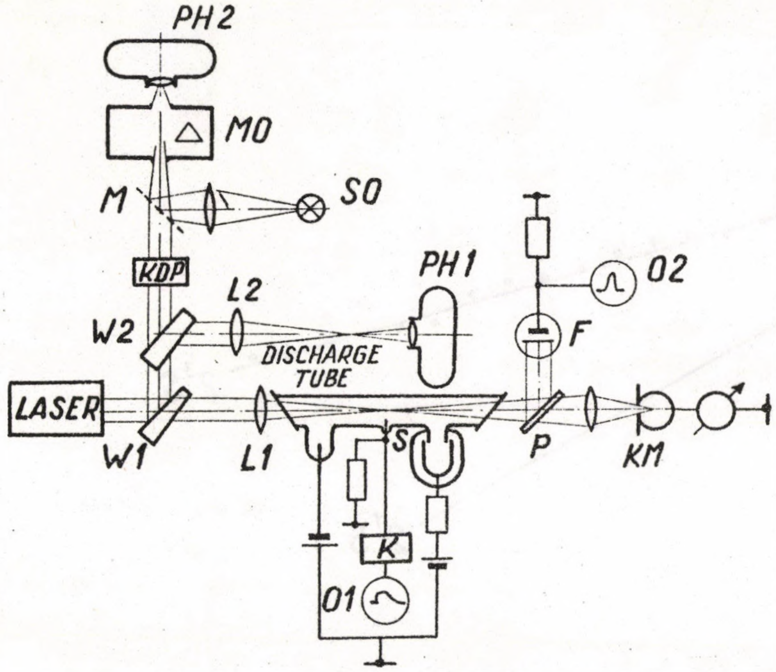


Fig. 13

Experimental set-up for the measurement of the five-photon ionization of triplet metastable He atoms in a glow discharge. The energy and time and spatial distributions of the laser pulse were measured with calorimeter KM, by photocamera PH1, in the focal volume of lens L2 /similar to lens L1/, and by the fast photodiode F coupled to fast oscilloscope O2. The frequency of the radiation was measured after frequency doubling by KDP, using the 5350 Å line of a Tl light source /SO/ as the frequency standard. MO and PH2 are a spectrograph and camera, respectively W1, W2 are glas wedges, K amplifier of the Langmuire probe S. The multiphoton signal is registered by oscilloscope O1.

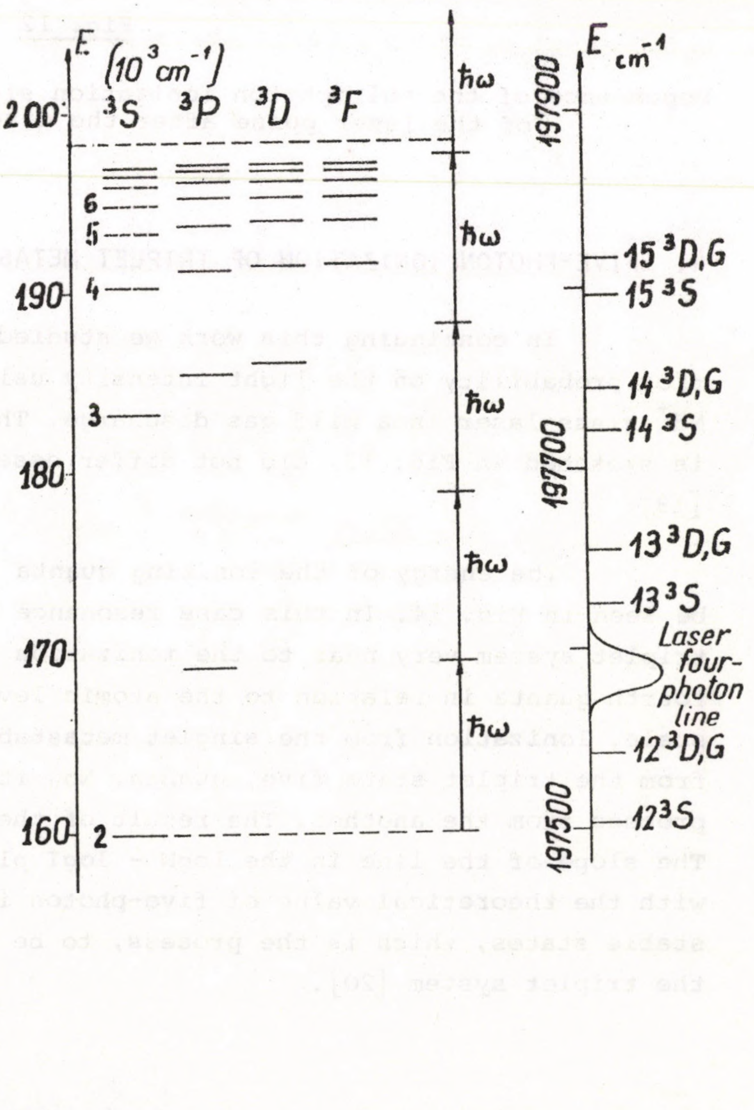


Fig. 14

Term schema of the triplet He atom with the energy of the light quanta of Nd glass laser radiation. There is four-photon resonance with the atomic states plotted in enlarged energy scale on the right-hand side of the figure

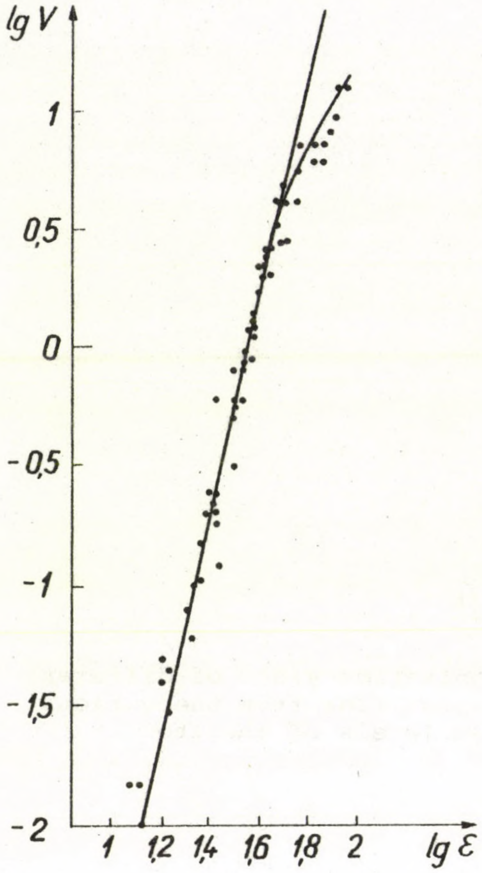


Fig. 15

Dependence of the five-photon ionization signal of the Langmuire probe on the energy of the laser pulse in the ionization of triplet metastable He atoms

Measurement of the multiphoton ionization in glow discharge nevertheless has some peculiarities in comparison to the measurement in the afterglow |19|. When we investigated the dependence of the multiphoton ionization probability on the intensity in a somewhat wider range of light intensities a complicated function was observed /Fig. 16/.

In discussing the measured dependence it should be noted that atomic states of different energies are simultaneously populated in the discharge and, equally, multiphoton ionization of different orders of these excited states is expected. To explain the lowest part of the measured curve we suppose that the one-photon ionization of levels of principal quantum number greater than three take place with simultaneous saturation, because of the high light intensity, and accordingly observed value of the slope $0.4 < 1$. The middle of the curve represents the two-photon ionization probability of the states of principal quantum number three. Finally, the upper part of the curve shows the intensity dependence of the five-photon ionization probability of the triplet metastable state. Three- and four-photon ionization cannot be observed because of the small populations of the corresponding excited states /for instance, the 2^1S states/. The ground state is never ionized because the light intensity required is too high. This fact is demonstrated experimentally by the failure to observe excess ions when the discharge is turned off.

To investigate the resonance in the triplet system we tuned the laser frequency in the range of the upper and middle parts of the curve shown in Fig. 16 by rotating a F.P. interferometer, with plates set a small distance of 50 μ apart, in the laser resonator itself [21]. Two of the measured curves can be seen in Fig. 17. It is apparent that the lower part of the curves remains unmodified when the field-free detuning of the 13^3S

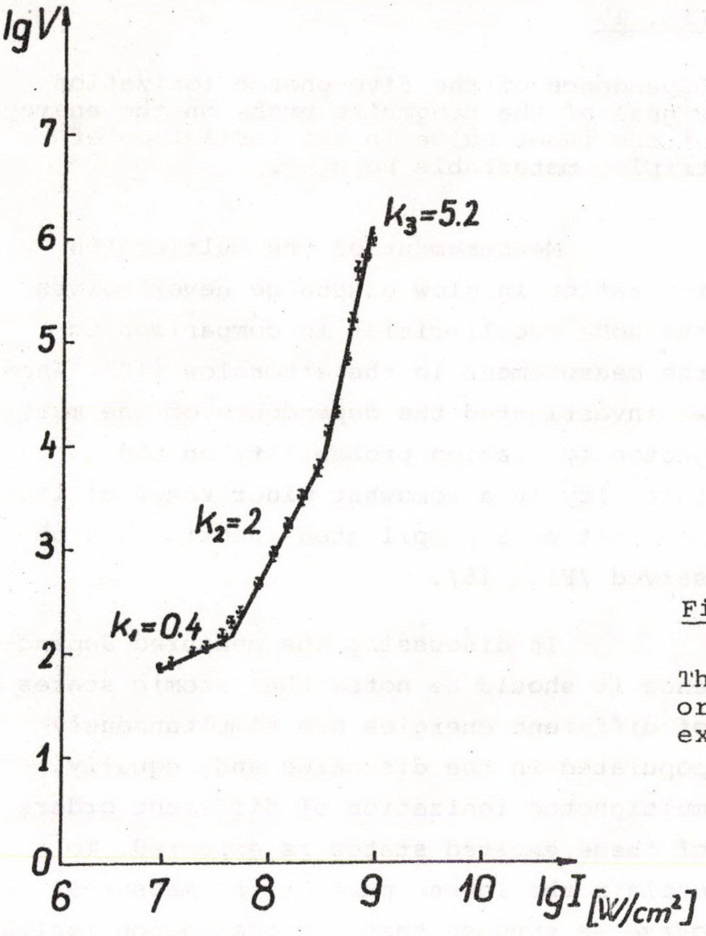


Fig. 16

The ionization yield of different order resulting from the various excited levels of the atom

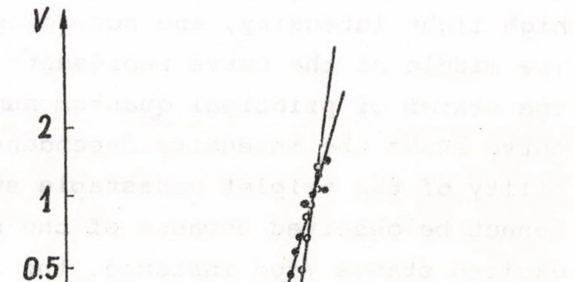


Fig. 17

Frequency dependence of the multiphoton ionization yield of the triplet metastable He atom. The parameter is the energy of the laser quanta in cm^{-1}.

level is altered by tuning the laser frequency, whereas the slope of the upper part of the curve changes drastically.

The novel feature of the measurement is that it represents the first case in which the slope k_{meas} has been observed to be greater than k_o. This drew our attention to the fact that the measured value can never be equated with the number of quanta absorbed in the ionization process, as had previously been supposed when only a fall in the slope of the line was observed. On plotting the value $k_{meas}-k = \Delta k'(4\bar{\nu})$ versus the energy of four quanta $4\bar{\nu}$ we obtain the curve of Fig. 18.

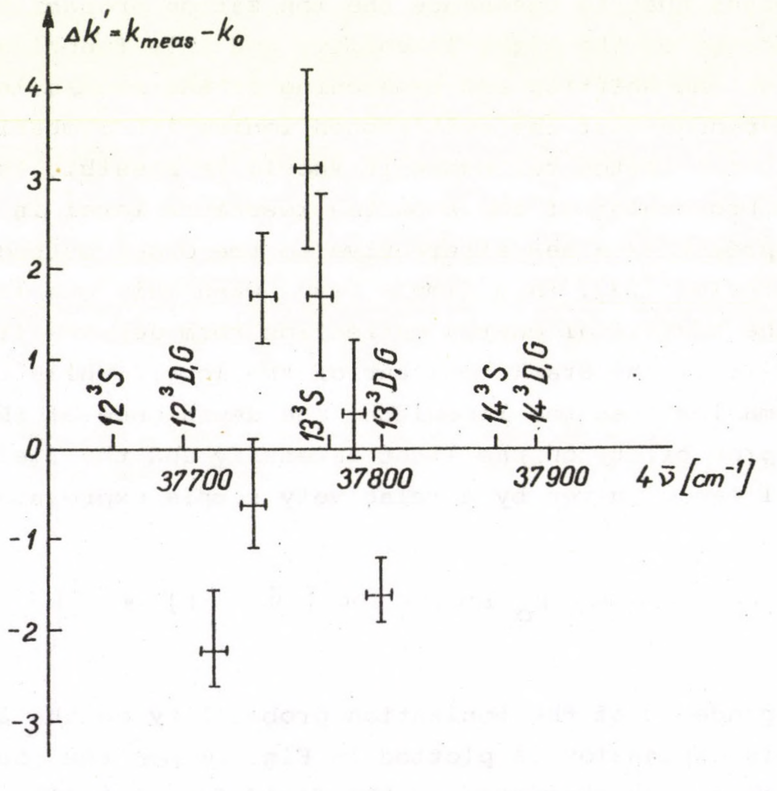


Fig. 18

Dependence of the deviation $\Delta k'$ of the factor k from the theoretical value k_o on the energy of four laser quanta

The result can be explained on the basis of the theory of multiphoton resonances described in the first section of the paper. Another interesting feature of the curve is the contrast between its dispersion-like form and the absorption-like form previously measured. The cause of this difference is that, relative to the energy of the laser quanta /9430cm^{-1}/, the resonance state of the case we are considering - the 13^3S state - lies at a smaller energy distance /600 cm^{-1}/ from the ionization threshold than the resonance levels hitherto discussed in the literature and therefore it has a smaller broadening and larger Stark shift. As a consequence the width of the resonance is determined only by the four-photon effective laser linewidth which is small in comparison with the Stark shift.

Detailed examination of the experimental curves, especially in the case of high values k_{meas}, shows that fitting of the curves on the assumption a linear relationship between the logarithm of the probability and the logarithm of the light intensity is not permissible [22]. This situation is what would be expected on the basis of expressions /4 and 5/ in the case of a high Stark constant and a comparatively wide range of the light intensity.

This means that in resonance the ionization probability becomes a complicated function of the light intensity, and this function contains information about the shifting and broadening of the atomic levels. Thus by measuring the dependence of the multiphoton ionization probability on the light intensity in k-photon resonance it should be possible to determine the Stark shift and broadening of the k-photon resonance level in the light field. Clearly this approach is a new alternative to the older method of measuring one-photon absorption [23]. In a simple case, when only one level is in resonance and the additional energy correction term depends linearly on the light intensity /c is the Stark constant of the level/ while the ionization broadening is smaller than the linewidth, the dependence of the logarithm of the ionization probability on the light intensity and the field-free detuning of the level can be given by a relatively simple expression:

$$\log W \sim k_o \log I - \log\left[(\Delta\bar{\nu} - cI)^2 + \gamma^2\right]$$

The dependence of the ionization probability on the light intensity according to this expression is plotted in Fig. 19 for the indicated numerical values of γ and c; the parameter is the field-free detuning $(\Delta\bar{\nu})$. It should be noted that the probability displays a "local maximum" at large detuning of, for instance, the laser frequency. The probability normally depends on the detuning of the laser frequency at low light intensity /before the cross points of the curves/; in other words, the smaller the detuning the larger the probability. But at high intensities this dependence is reversed.

The two curves of Fig. 20, measured at different values of field-free detuning, clearly resemble the theoretical curves plotted in Fig. 19. It is apparent that the 13^3S state is shifted into resonance down from the ionization threshold, because the higher the field-free detuning the larger is the light intensity required to tune the level into resonance. The mark on the axis indicates the light intensity where the level is in perfect resonance with the energy of four quanta.

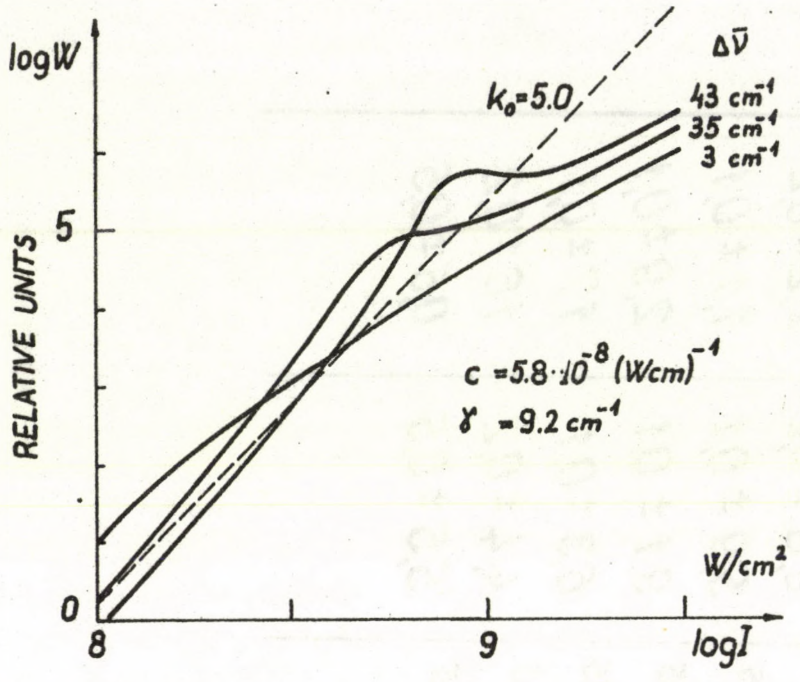


Fig. 19

Theoretical dependence of the multiphoton ionization probability on the light intensity taking into account the shift of the resonance level under the influence of the ionizing radiation. The parameter is the field-free resonance detuning

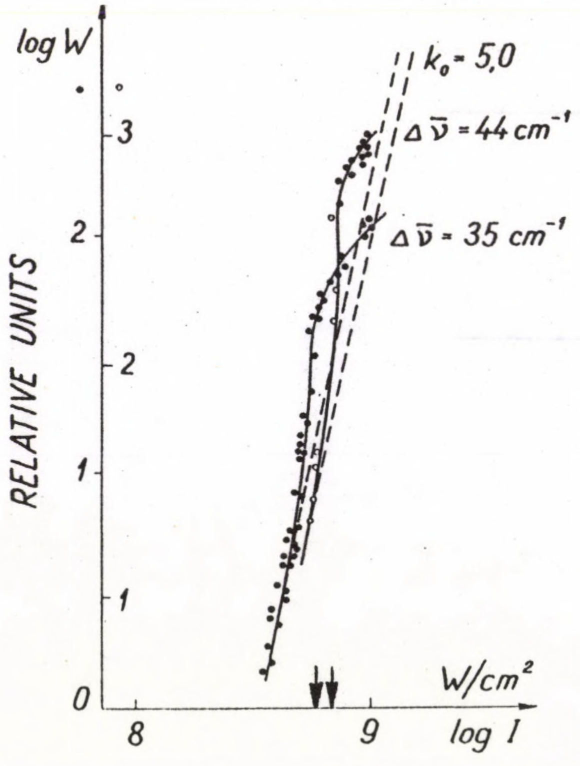


Fig. 20

The experimentally measured dependence of the multiphoton ionization probability on the light intensity at different values of field-free detuning

TABLE I.

Atom	$\hbar\omega$ eV	K_0	E V/CM	K	$K_0 - K$
Xe	1,18	11	$10^{7,66 \pm 0,15}$	8,8 ± 0,2	2,2 ± 0,2
Xe	1,78	7	$10^{7,55 \pm 0,15}$	5,9 ± 0,1	1,1 ± 0,1
Kr	1,18	12	$10^{7,71 \pm 0,15}$	9,1 ± 0,1	2,9 ± 0,1
Kr	1,78	8	$10^{7,55 \pm 0,15}$	6,3 ± 0,1	1,7 ± 0,1
Xe	2,36	6	$10^{7,34 \pm 0,15}$	4,4 ± 0,2	16 ± 0,2
Kr	2,36	6	$10^{7,34 \pm 0,15}$	5,5 ± 0,5	0,5 ± 0,5

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