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RESONANCE MULTIPHOTON IONIZATION

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RESONANCE MULTIPHOTON IONIZATION

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SUMMARY:

The five-photon ionization of metastable helium atoms was investigated in the case of four-photon resonance with atomic levels lying in the energy range close to the ionization limit, where the states are closely packed. The dependence of the five-photon ionization yield on the light intensity and frequency was measured. Using the experimental results, the distortion of the atomic structure in light fields of high intensity/i.e. the dependence of atomic term energy on the light intensity/ was determined. Some limitations to the applicability of perturbation theory were revealed.

KIVONAT

Metastabil hélium atomok ötfotonos ionizációját vizsgáltuk négyfotonos rezonancia esetén. Mértük az ötfotonos ionizáció valószinüségének függését a fény intenzitásától és frekvenciájától. A kisérleti adatok felhasználásával meghatároztuk az atomi energianivók termékének a fény intenzitásától való függését, ezáltal meghatároztuk az atomi szerkezet torzulását erős fényterekben. A mérési adatok egy része azt mutatta, hogy a jelenségek meghatározott körében a perturbációszámitás már nem alkalmazható.

РЕЗЮМЕ

Пятифотонная ионизация метастабильных атомов гелия изучалась в случае четырехфотонного резонанса с атомными состояниями, расположенными в области энергии, близкой к границе ионизации и, следовательно, тесно примыкающими друг к другу. Измерялась зависимость вероятности многофотонной ионизации от интенсивности и частоты лазера. По результатам эксперимента определялась энергия атомных состояний в зависимости от интенсивности света, что позволяет судить об искажении атомной структуры в сильном поле излучения и оценить границу применимости теории возмущений.

I. INTRODUCTION

The dependence of the rate of multiphoton ionization W on the light intensity /I/, in the approximation of the first non vanishing order k_o of the perturbation theory, is

$$W = \alpha I^{k_o} \qquad /1/$$

where

$$k_o = < \frac{IP}{\hbar\omega} + 1 > \qquad /2/$$

is the number of photons absorbed in the ionization process. IP is the ionization potential of the initial state /i/, $\hbar\omega$ is the energy of the ionizing quantum, and the "cross section" of the ionization

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

is constant.

The density /i.e. yield/ of ions /N_i/ produced by multiphoton ionization is proportional to this probability W:

$$\log N_i \sim \log W \sim k_o \log I \qquad /4/$$

Now in many cases the experimentally measured slope, k, of the $\log N_i$ vs. logI plot turns out to be smaller than the theoretical value, k_o, especially in the ionization processes of the rare gases [1,2,3]. In explaining this fact states near to the ionization limit of the atom are supposed to be smeared under the influence of high-intensity ionizing light /continuum like spectrum/, the result of which is an apparent fall in the ionization potential of the initial state [4].

Another cause for decrease of the factor k may be multiphoton

resonance phenomena [5]. Such a decrease has been demonstrated experimentally by the Delones and their co-workers for the case where the energy of $k < k_o$ quanta coincides with the energy of an atomic state [6]. The approximation of k_o order in the perturbation theory, it is found, does not satisfactorily describe this process, and allowance must be made for approximations of higher order [7,8]. In the case of resonance one of the denominators of expression /3/ tends to zero and the dependence of the energy of the states on the intensity in this denominator has to be taken into account. Hence, supposing

$$E_j - E_o - k\hbar\omega + i\gamma_j \to 0 \qquad /5/$$

then

$$E_j = E_{jo} + \Delta E_j(I) \qquad /6/$$

$$E_o = E_{oo} + \Delta E_o(I) \qquad /7/$$

$$\gamma_j = j_{jo} + \Delta\gamma_j(I) \qquad /8/$$

E_{jo} and E_{oo} are the field-free energies of the j-th state and ground state, while γ_{jo} is the field-free level width of the j-th state. Consequently, the "cross section" ceases to be constant and the multiphoton ionization rate becomes a complicated function of the light intensity / W/I/ /, which in turn is a function of the light frequency / $W = W/I,\bar{\nu}$/ /.

In practice this complex function is approximated over small ranges of light intensity by the linear function in the logW-logI plot, i.e. by the tangent of the logarithm of the multiphoton ionization rate-light intensity function. The slope of this line is the measured value of the factor [5]

$$k(I_o,\bar{\nu}) = \left(\frac{\partial \log W(I,\bar{\nu})}{\partial \log I}\right)_{I=I_o} \lesseqgtr k_o \qquad /9/$$

Experimentally only values of $k/I_o,\bar{\nu}$/ smaller than, or equal to, k_o had been observed before our own work in Budapest.

2. SETTING OF THE PROBLEM

The aim of this work was basically twofold:

1/ to study the detailed features of multiphoton resonances and their role in multiphoton ionization; and

2/ to investigate the behaviour of atomic states near to the ionization limit of the atom under the influence of high-intensity ionizing radiation and the effect of these on the multiphoton ionization rate.

For this purpose we chose to study the multiphoton ionization process from the triplet metastable state of the He atom, using the radiation of a Q-switched, frequency-tuned Nd^{3+} glass laser. This ionization process is a five-photon ionization with four-photon resonances of the n^3S, D, G states, where $10 < n < 16$. These states lie at an energy distance of about 600 cm^{-1} from the ionization limit of the atom, and the frequency of the laser light can be tuned into resonance with each of term in succession.

This type of resonance multiphoton ionization is especially interesting because a large decrease was observed in the nonlinearity of multiphoton ionization /k/ from the ground state [9]. The four-photon resonance states of five-photon ionization from the triplet metastable state of helium fall within the energy range of level smearing of the multiphoton ionization process from the ground state. It was therefore - expected as it turned out, correctly - that we should be able to observe simultaneously both the beginning of the process leading to the smearing of levels in the relatively weak light field of the five-photon ionization and the role of the resonance in the energy region close to the ionization limit. These observations have enabled us to make fairly definite conclusions about the limit of applicability of the perturbation method for the description of the features and mechanism of the multiphoton ionization process.

3. EXPERIMENTAL SET-UP

The experimental set-up that was employed can be seen in the Fig.1. The laser was Q-switched with a rotating prism and frequency-tuned in the laser cavity with an excess F.P. interferometer. The parameters of the laser radiation /farfield pattern, intensity-time function, pulse energy, etx./ were measured in the usual way [13] in order to calculate the peak light power [10]. The laser frequency was measured shot by shot in absolute scale after frequency-doubling by KDP. The helium atoms were excited to the metastable state in the glass tube by mild gas discharge and the ions were

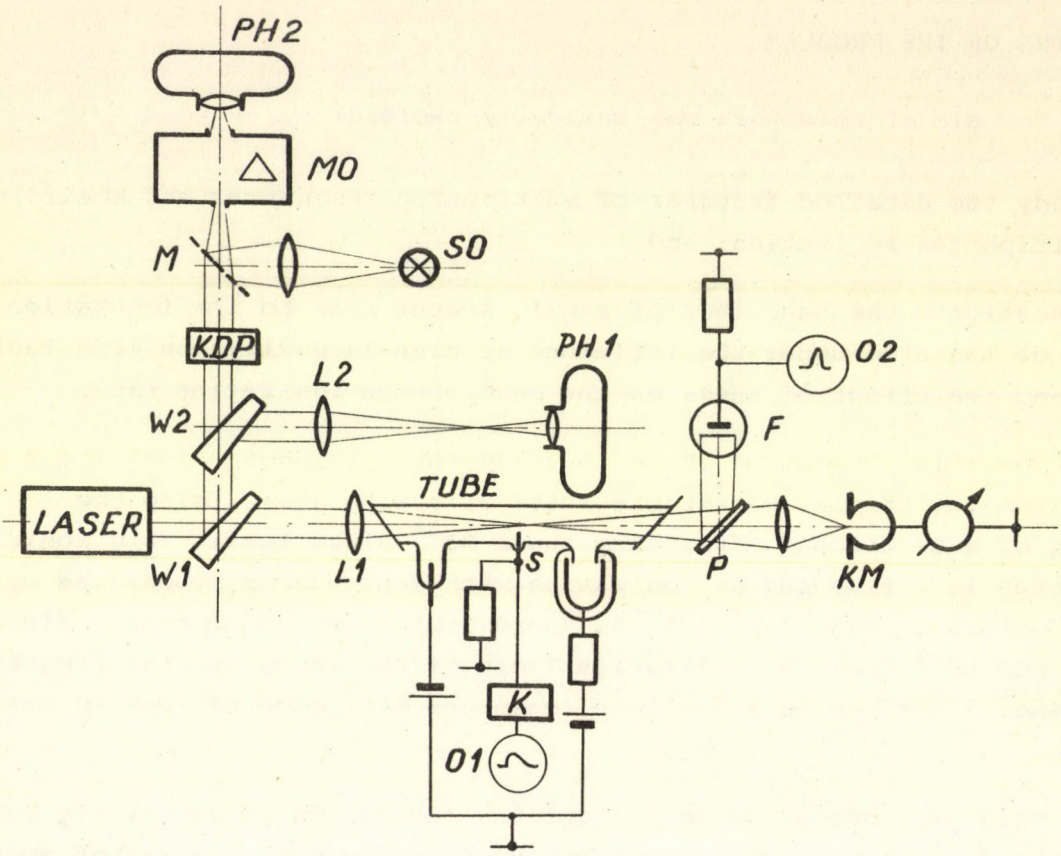


Fig. 1

detected with a Laingmuire probe [11]. The amplitude of the pulse of the Langmuire probe /V/ is proportional to the maximum ion density induced by the focused laser radiation [12].

4. IONIZATION OF DIFFERENT ORDER

The peculiar feature of measuring multiphoton ionization yield with this experimental set-up is that multiphoton ionizations of different order occur parallelly, because states of different ionization potential are simultaneously populated by the discharge [13]. The term scheme of the triplet He system is plotted on Fig. 2. From states lying in the zone (1) one-photon ionization takes place; from the states of the second zone (2) two-photon ionization, and so on. Because of the variation in "cross section" of these different-order processes and their dissimilar power dependences on the light intensity, it is possible to observe their separate contributions to the total yield of the ionization. The result of the measurement is plotted on Fig. 3. At the bottom of the figure can be seen the dependence of the one-photon ionization yield on the light intensity; $k = 0.4$ because of the saturation of the process. The middle

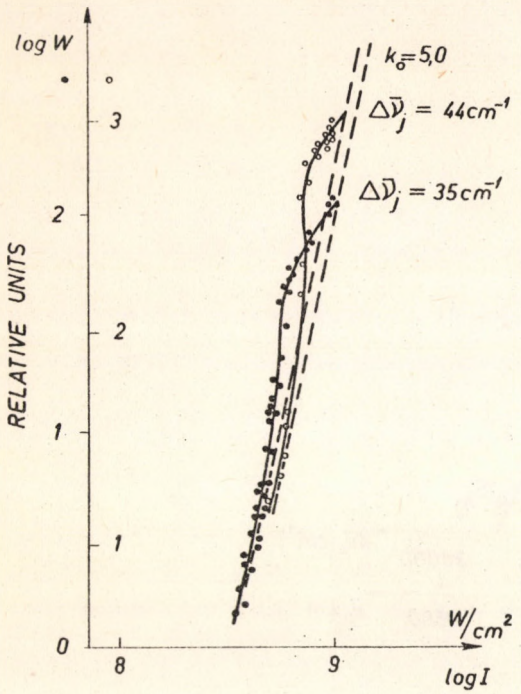


Fig. 12

It is apparent that the dynamical resonance intensity $/I_{Rj}/$ depends on the field-free detuning, i.e. on the frequency of the laser radiation in the following form

$$I_{Rj} = \frac{\Delta\bar{\nu}}{c_j - c_o} = \frac{E_{jo} - E_{oo} - 4\bar{\nu}}{c_j - c_o} \qquad /13/$$

As Fig. 13 demonstrates the value of I_R is the intensity corresponding to the intersection of the tangents before and after the resonance. These curves are theoretical lines plotted on the assumption that the resonance occurs at 1 GW/cm^2 intensity; the parameter is the laser linewidth. It is clear that the value of I_{Rj} determined by the above method slightly depends on the effective width of the resonance and therefore the dynamical resonance intensity obtained from the measured logW - logI curves can be considered to be the real one.

Using this method and the measured logW - logI curves, we can go on to plot the position of the resonance intensity $/I_{Rj}/\bar{\nu}//$ in the light intensity-frequency coordinate system. The result is presented in Fig. 14. This shows how the dynamical resonance intensity $/I_{Rj}/$ of the different atomic four-photon resonance states /j/ depends on the light frequency $\bar{\nu}$ /i.e. on the energy of four quanta $4\bar{\nu}$/. In the case of levels indicated on the right-hand side of the figure the smaller the light frequency the higher is the light intensity required to reach dynamical resonance. Consequently, if we suppose that only the four-photon resonance state is shifted $/c_o = 0/$, and if the energy is counted from the initial state, these lev-

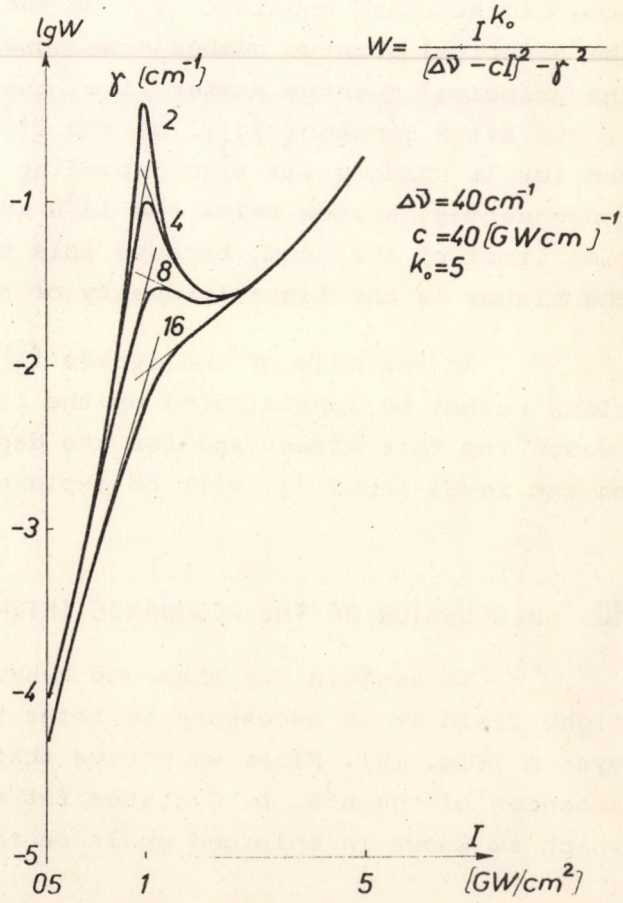


Fig. 13

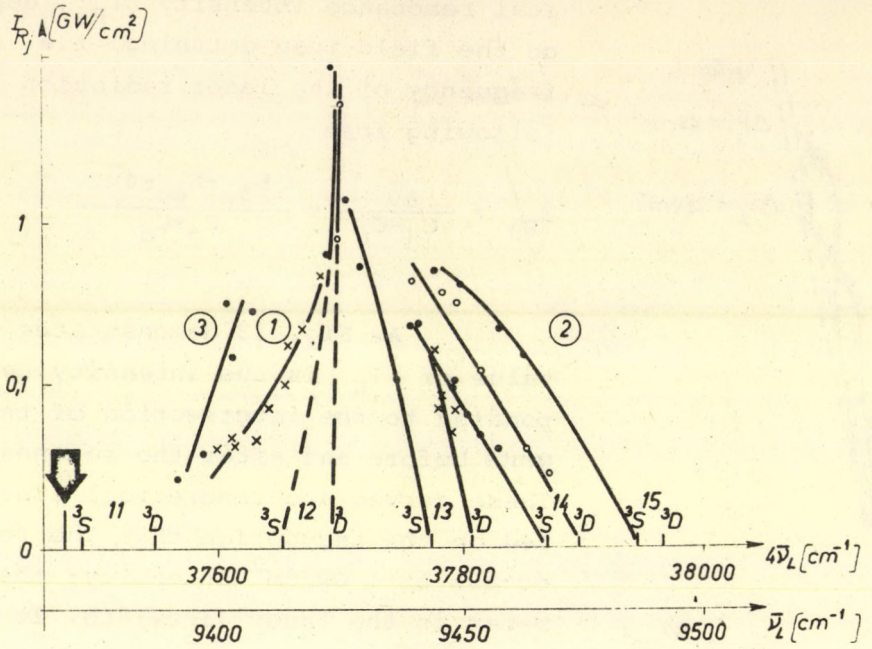


Fig. 14

els are shifted by the field of the radiation downwards from the ionization limit of the atom. The reciprocal value of the slope of the $J_{Rj}/4\bar{\nu}/$ line is the Stark constant of the level $/c'_j/$. Investigating the dependence of the Stark constant c'_j on the level index /j/ - for instance, on the principal quantum number - we conclude from Fig. 14 that the smaller the principal quantum number /i.e. the energy of the level /j//, the higher is its Stark constant $/c'_j/$. At the $13\,^3S$, D states the Stark constant of the levels changes its sign depending on the principal quantum number. Consequently, states below the $12\,^3D$ state shift upward towards the ionization limit of the atom, because this time the higher the light frequency, the higher is the light intensity of the dynamical resonance /see Fig. 14/.

In the case of the curves ① and ② of Fig. 14 the $J_{Rj}/\bar{\nu}/$ functions cannot be approximated by the linear relationship $/I_{Rj} \sim \frac{1}{c}, \bar{\nu}/$. The reason for this effect and for the dependence of the Stark constant $/c'_j/$ on the level index /j/ will be explained in the following section.

10. DISCUSSION OF THE RESONANCE INTERACTIONS

To explain the observed behaviour of the resonance states in the light field it is necessary to refer to the term scheme of the He triplet system /Fig. 15/. First we notice that over and above the four-photon resonances of the n^3S, D, G states /fat arrows ① ② ③/, the energy of which is shown in enlarged scale on the right-hand side of the figure,

width /γ/, the dynamical resonance detuning /i.e. the resonance denominator in expression /9// is given by

$$E_{jo} - E_{oo} + (C_1 - C_o)\ I_{rj} - 4\bar{\nu} = 0 \qquad /11/$$

$\hbar, c = 1$ is supposed.

7. GENERAL NONLINEARITY FUNCTION

By measuring the logW - logI function we can establish the nonlinearity of the processes only as the slope of the tangent of the plot at some definite light intensity /I_1/. The nonlinearity becomes a fast-varying function of the light frequency and intensity.

To illustrate this the plot of the measured nonlinearity of five-photon ionization of the triplet metastable helium atoms is shown in Fig. 10 as a function of the frequency. Unfortunately, it is difficult to measure this fast-varying function to the desired accuracy, especially as far as determining the frequency is concerned. /The estimated accuracy of the frequency measurement was 0.4 cm^{-1}./ Consequently, it is hard to identify with certainty the state that is in resonance and hence to deduce from the function any information about the energy of the resonance state and its change under the influence of the laser field.

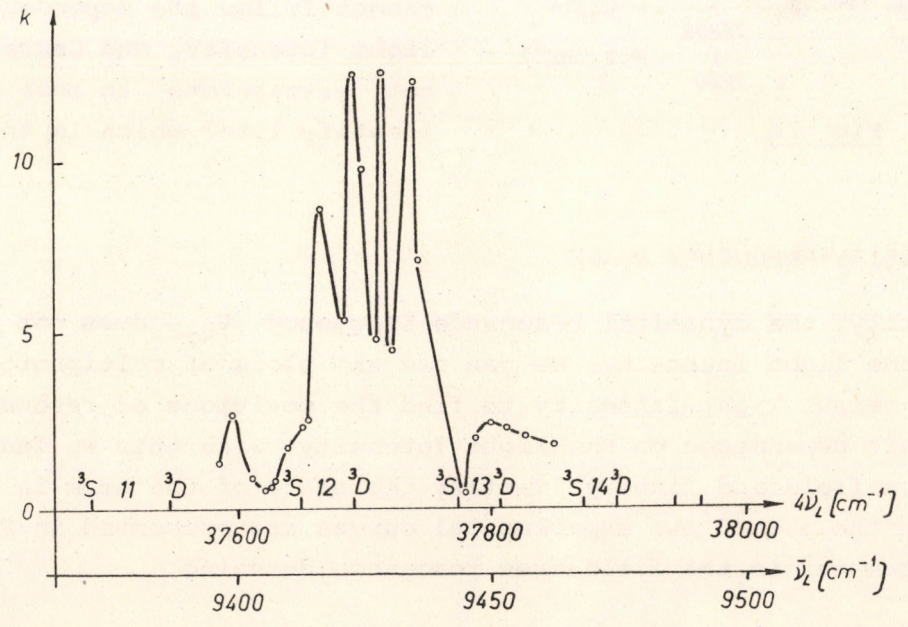


Fig. 10

8. DISPERSION CURVE

There is however, a further possible source of information, namely, the dispersion curve of the multiphoton ionization rate [18], or in other words, the dependence of this rate on the light frequency at fixed light intensity / $W/\bar{\nu};I_1$ // because this function reaches a maximum where the dynamical detuning is zero.

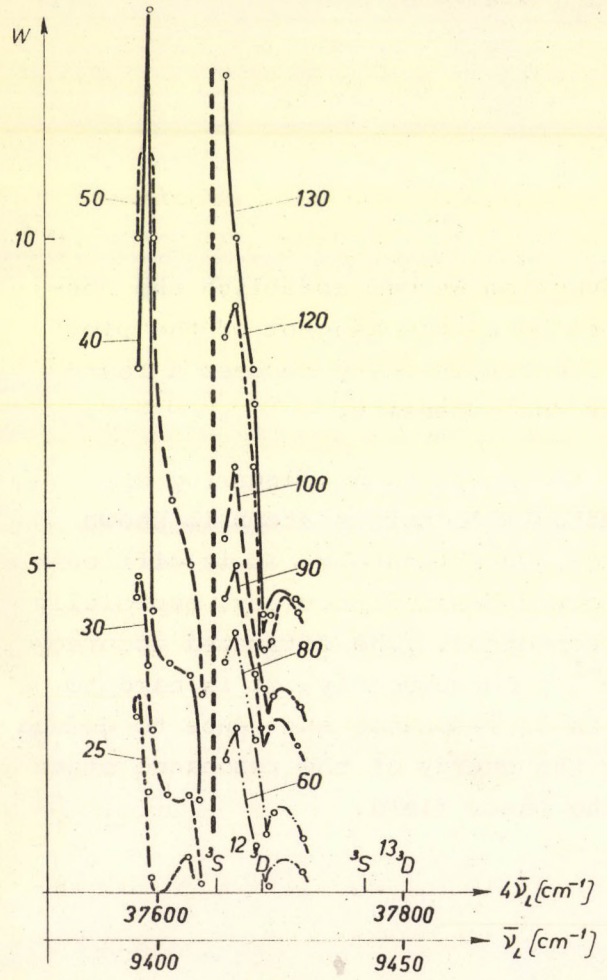


Fig. 11

On Fig. 11 we have plotted the measured dispersion curve of five-photon ionization of the triplet metastable helium atom at different light-intensities. Once again, though, it is evident that we face the same difficulty as in the case of the nonlinearity-frequency function. The multiphoton ionization rate depends strongly on the light frequency, and the accuracy of the frequency measurement is not sufficient to allow us to determine the exact position of the maximum. This means that we cannot follow its dependence on light intensity, and hence we cannot extrapolate to zero field to identify level which is in resonance.

9. THE INTENSITY-FREQUENCY PLANE

Luckily, the dynamical resonance frequency ∇_{Rj}, does not depend strongly on the light intensity. We can use the plots of multiphoton ionization rate versus light intensity to find the positions of resonances and to follow their dependence on the light intensity, with this we can extrapolate to zero field and finally identify the state of the atom in resonance. To illustrate the method two experimental curves are presented in Fig. 12 [19]; the parameter is the field-free resonance detuning

$$\Delta\nu_j = E_{jo} - E_{oo} - 4\bar{\nu} \qquad /12/$$

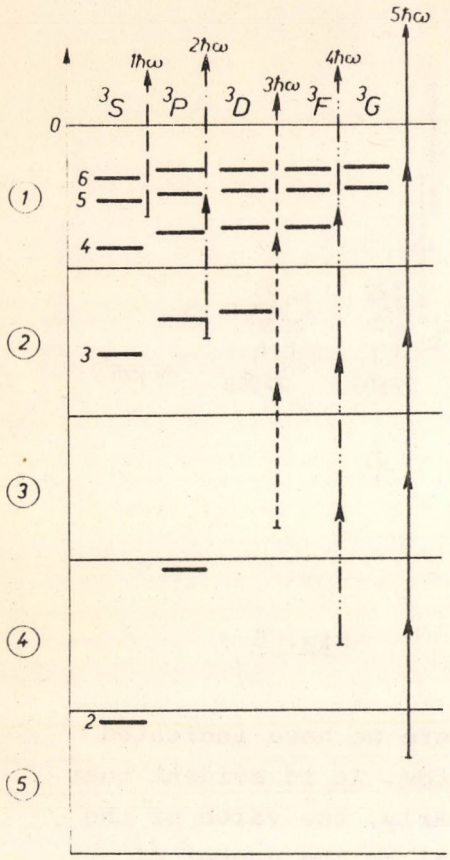


Fig. 2

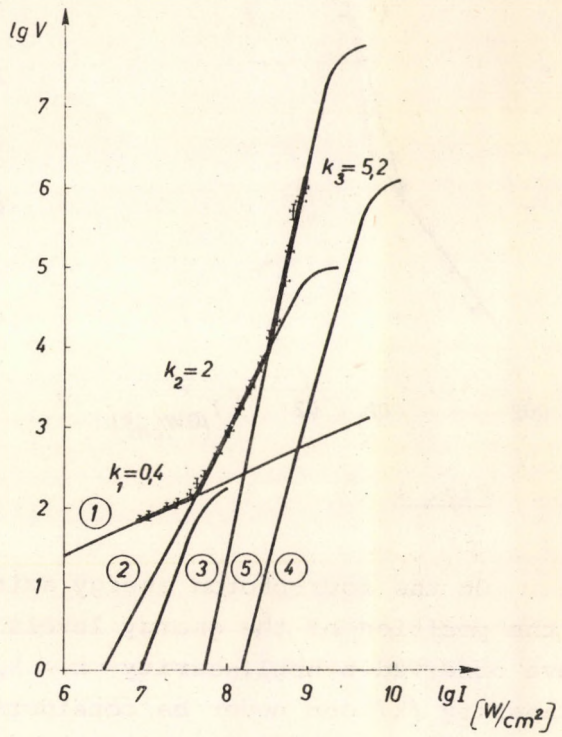


Fig. 3

section of the curve shows the contribution of two-photon ionization to the total ionization yield, while the upper section represents the five--photon ionization yield from the triplet metastable state.

5. NONLINEARITY

To investigate the influence of four-photon resonances on the five-photon ionization yield the frequency of the laser radiation was varied and the ionization yield versus light intensity function /$N_i \sim W/I;\bar{\nu}$/ was measured at different light frequencies [14]. Two typical curves are plotted in Fig. 4. It is apparent that, to a first approximation, the contribution of two-photon ionization is independent of the light frequency, whereas the five-photon ionization yield changes drastically. Adopting the usual linear approximation in the logW - logI coordinate system, we can plot the frequency dependence of the nonlinearity of the process /i.e. the value of the slope of this line /$k/\bar{\nu};I_1$/ /in logW - logI plot/ at constant light intensity /$I_1 = 2.10^8$ Wcm^{-2}/ /Fig. 5/.

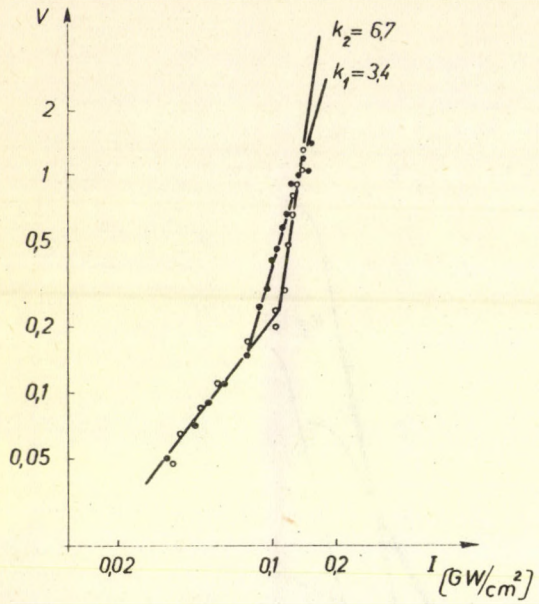


Fig. 4

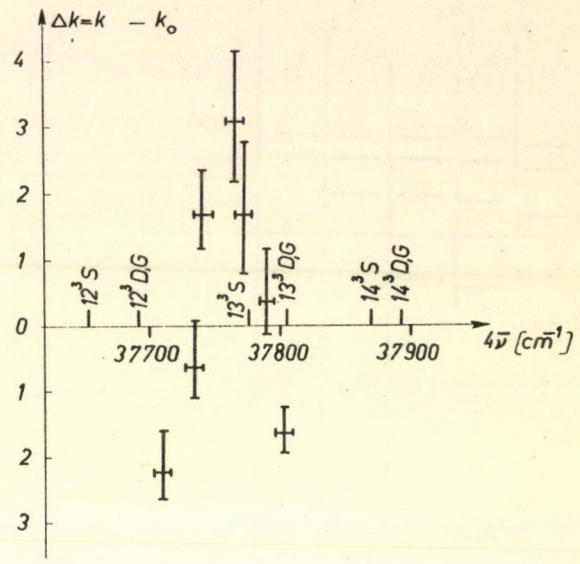


Fig. 5

On the four-photon energy axis of the figure we have indicated also the position of the energy levels of the He atom. It is evident that we have observed a nonlinearity $k > k_o$, so, clearly, the value of the nonlinearity /k/ can never be considered to be equal to the number of quanta absorbed during the ionization; this variation of the factor k with light intensity is a feature of the approximation of higher than k_o order, i.e. the consequence of a shift of levels in the high-intensity laser field. We should further note the fast change of the nonlinearity with frequency, which points to the existence of distinct atomic states even in the energy range close to the ionization limit.

6. GENERAL RATE VS. INTENSITY FUNCTION - STATISTICS - LINE SHAPE

During subsequent measurements it became obvious that the linear approximation in the logW - logI coordinate system is simply inadequate for describing the multiphoton ionization rate-light intensity dependence in the resonance case and over a wide range of light intensities [15]. We postulate that the required energy correction term $/\Delta E_j/I//$ to the four--photon resonance state /j/ depends linearly on the light intensity:

$$\Delta E_j(I) = C_j\, I \qquad /10/$$

The dependence of the five-photon ionization rate on light intensity $W/I;\bar{\nu},\gamma/$ resulting from expression /3/ on the assumption that the level is

in four-photon resonance is plotted for the different level widths /γ/ in Fig. 6.

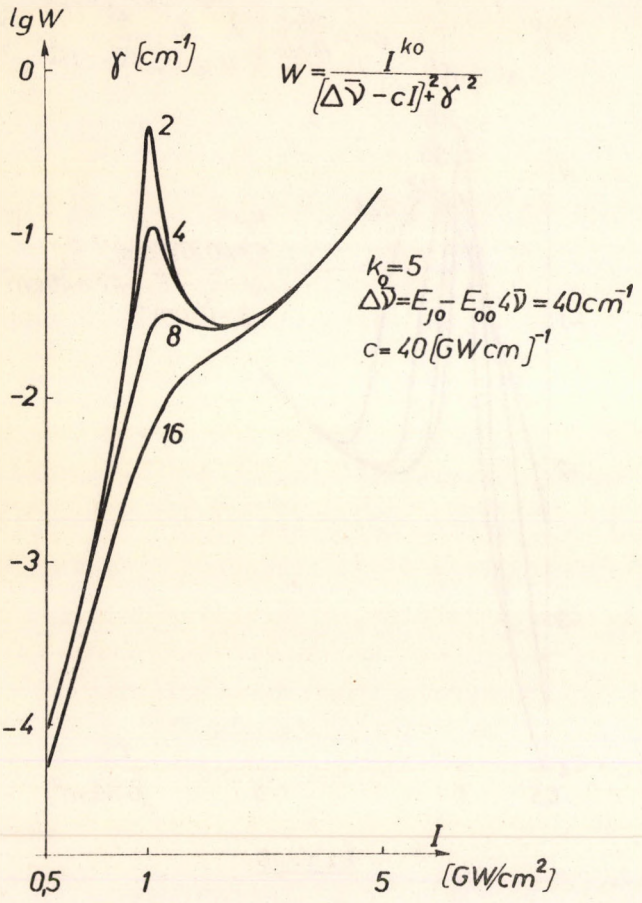


Fig. 6

In our measurement we used a laser working in multimode operation and therefore the laser could be considered as being a thermal light source. Agarwal [16] has shown that if the laser line shape is Lorentzian, so also will be the resultant dependence of the multiphoton ionization rate on the frequency, and the resultant level width $\gamma_r = N\gamma_L + \gamma$. Therefore, provided the assumption we have made are correct, the curves presented in Fig. 6 should describe the real situation with $\gamma = \gamma_r$. Given that the shape of the atomic level is always Gaussian, owing to Doppler effect, we obtain the curves shown in Fig. 7. These do not deviate strongly from the curves of Fig.6, because the Doppler level width /ΔE_D/ is always smaller than the laser linewidth /γ_L/.

The supposition of a Gaussian-shaped laser line /with linewidth $\Delta\bar{\nu}$/, on the other hand, yields curves of W/I;$\bar{\nu}$,$\Delta\bar{\nu}$/ which deviate very considerably from the previous curves, as can be seen from Fig. 8. Furthermore, the supposition of Gaussian atomic level and Gaussian laser line, a case which was calculated by Rapoport et al. [17] in connection with the multiphoton absorption process, leads to almost identical curves.

The logW - logI function actually measured by us is presented in Fig. 9. The solid line was plotted on the assumption that the laser line is of Lorentzian shape. It is readily apparent that the experimental curve cannot be described by the supposition of a Gaussian laser line because of the large differences between it and the theoretical curve /cf. Fig. 8 and 9/.

Exact dynamical four-photon resonance occurs at the intensity $I_r = 6.10^8$ W/cm^2; that is, at this intensity, and neglecting the level

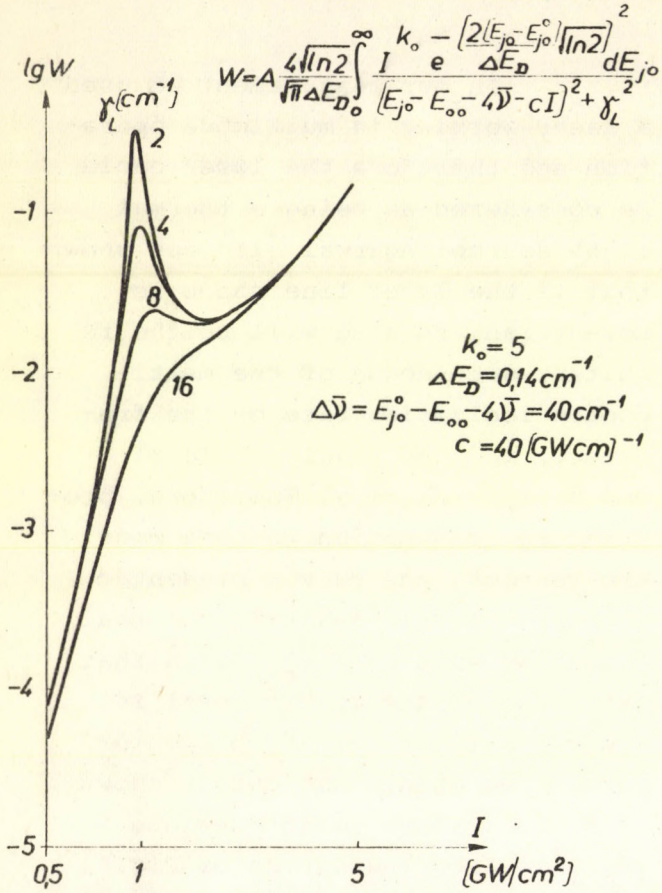


Fig. 7

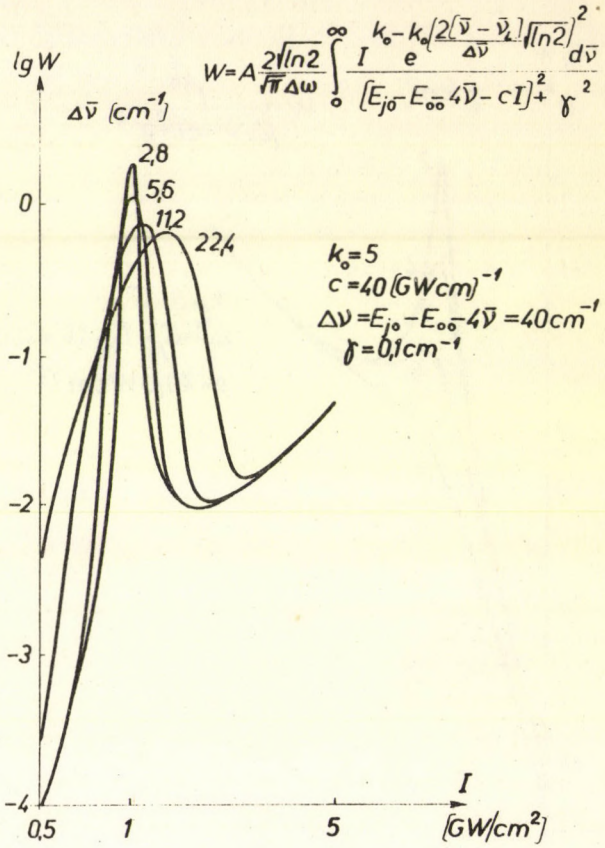
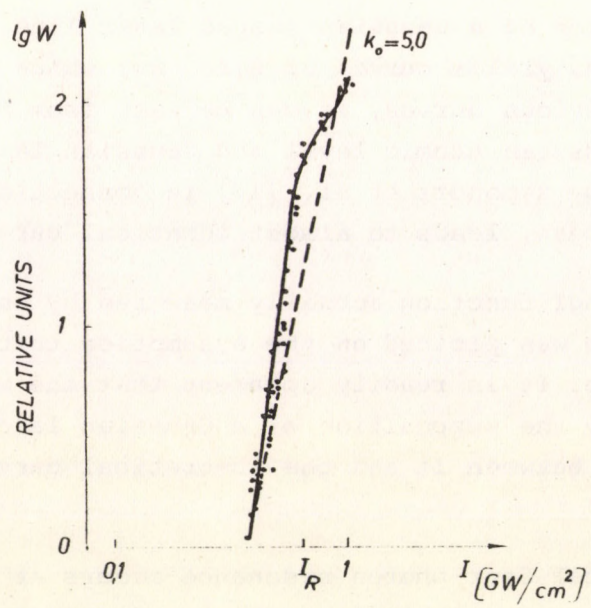


Fig. 8



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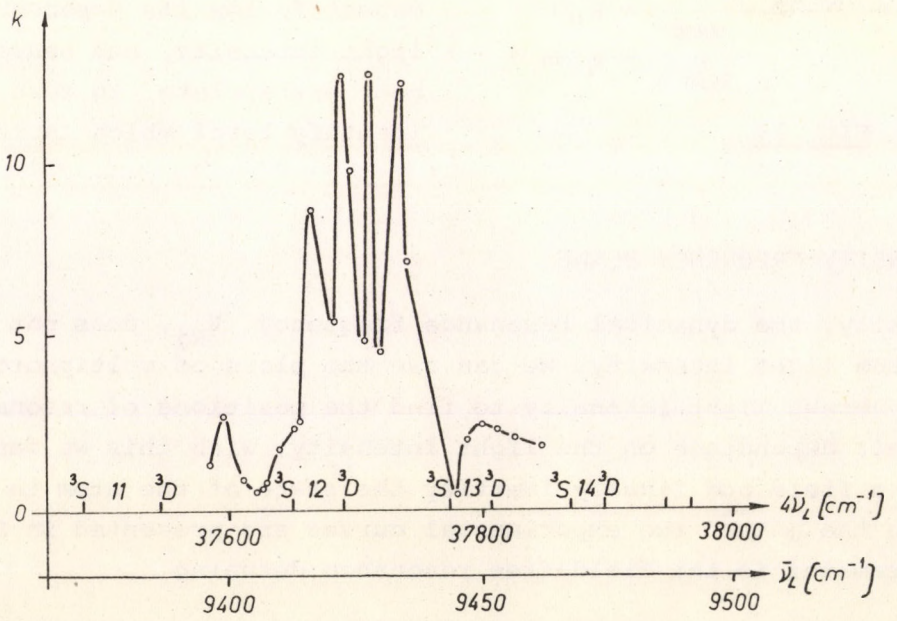

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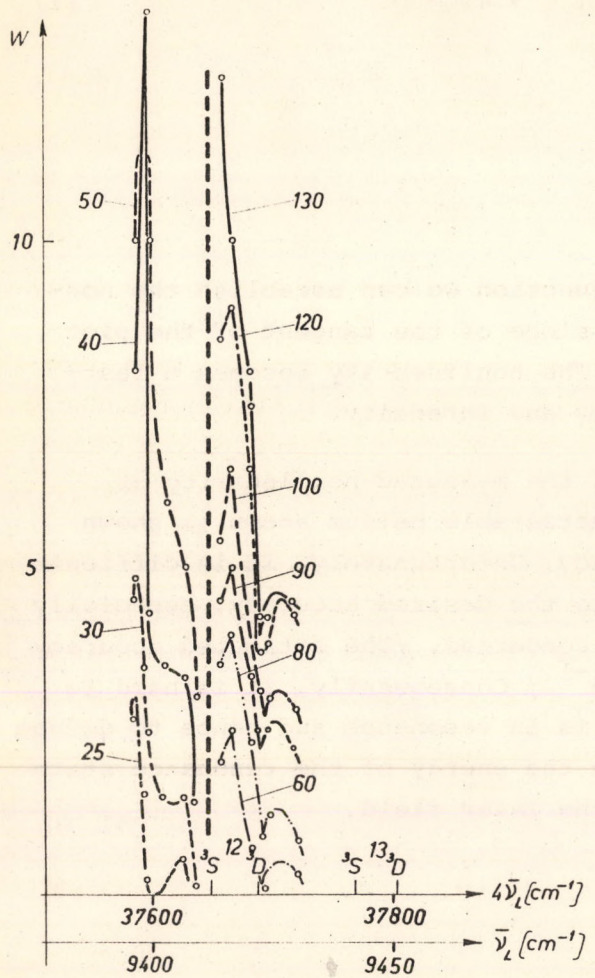

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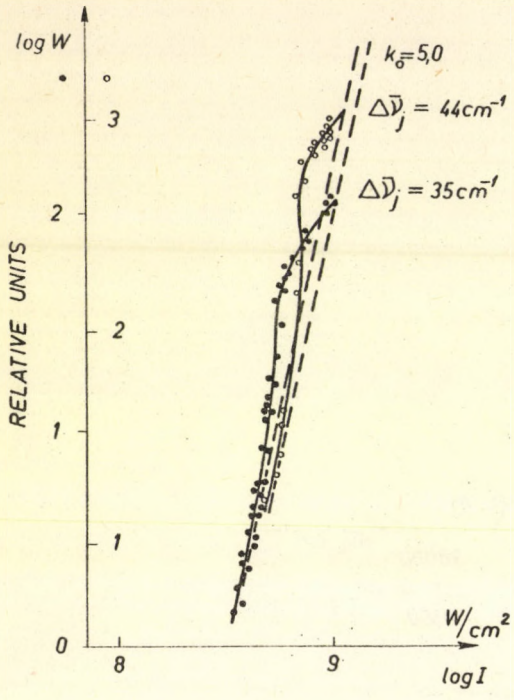

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$$I_{Rj} = \frac{\Delta\bar{\nu}}{C_j - C_o} = \frac{E_{jo} - E_{oo} - 4\bar{\nu}}{C_j - C_o} \qquad /13/$$

As Fig. 13 demonstrates the value of I_R is the intensity corresponding to the intersection of the tangents before and after the resonance. These curves are theoretical lines plotted on the assumption that the resonance occurs at 1 GW/cm^2 intensity; the parameter is the laser linewidth. It is clear that the value of I_{Rj} determined by the above method slightly depends on the effective width of the resonance and therefore the dynamical resonance intensity obtained from the measured logW - logI curves can be considered to be the real one.

Using this method and the measured logW - logI curves, we can go on to plot the position of the resonance intensity $/I_{Rj}/\bar{\nu}//$ in the light intensity-frequency coordinate system. The result is presented in Fig. 14. This shows how the dynamical resonance intensity $/I_{Rj}/$ of the different atomic four-photon resonance states /j/ depends on the light frequency $\bar{\nu}$ /i.e. on the energy of four quanta $4\bar{\nu}$/. In the case of levels indicated on the right-hand side of the figure the smaller the light frequency the higher is the light intensity required to reach dynamical resonance. Consequently, if we suppose that only the four-photon resonance state is shifted $/c_o = 0/$, and if the energy is counted from the initial state, these lev-

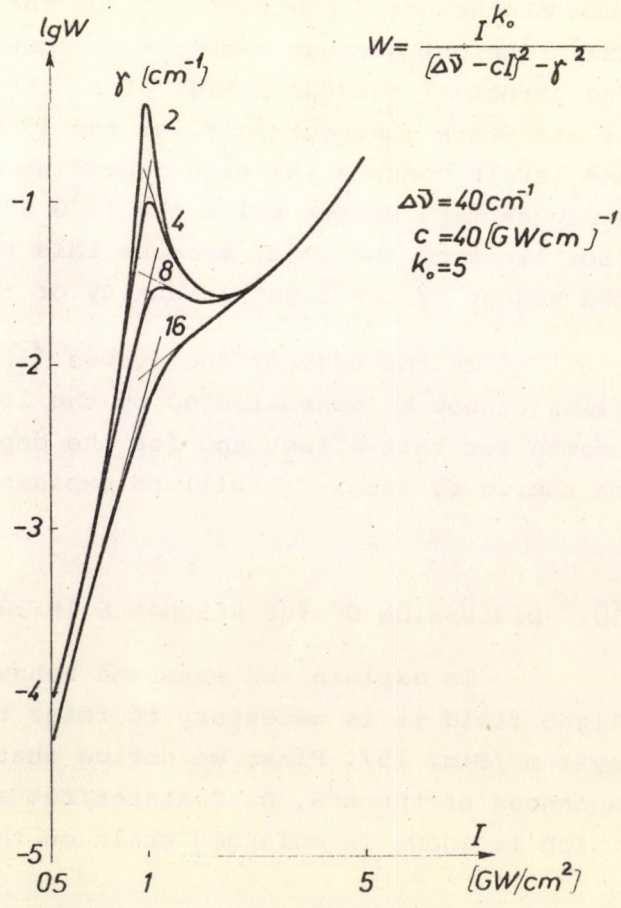

Fig. 13

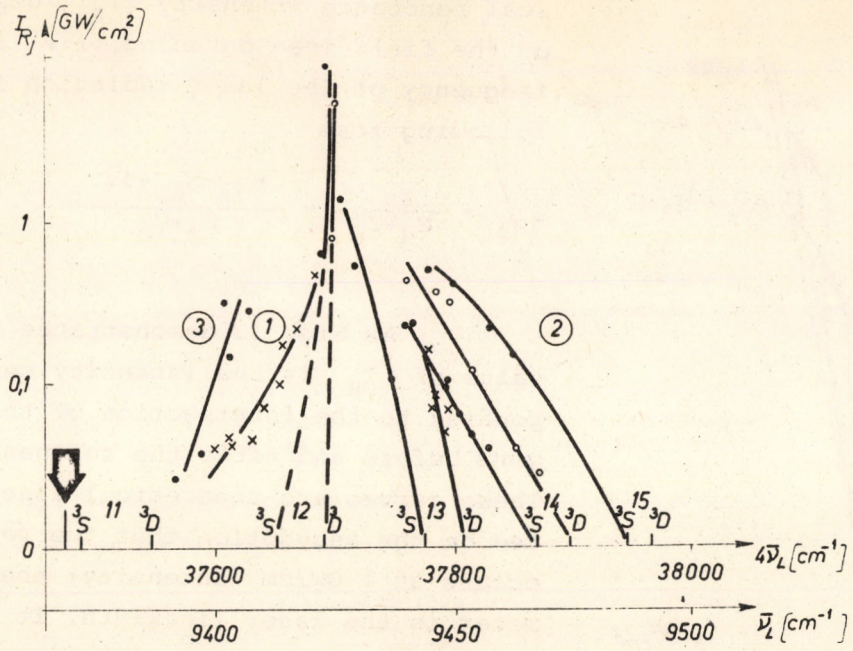

Fig. 14

els are shifted by the field of the radiation downwards from the ionization limit of the atom. The reciprocal value of the slope of the $J_{Rj}/4\bar{\nu}/$ line is the Stark constant of the level $/c'_j/$. Investigating the dependence of the Stark constant c'_j on the level index /j/ - for instance, on the principal quantum number - we conclude from Fig. 14 that the smaller the principal quantum number /i.e. the energy of the level /j//, the higher is its Stark constant $/c'_j/$. At the $13\,^3S$, D states the Stark constant of the levels changes its sign depending on the principal quantum number. Consequently, states below the $12\,^3D$ state shift upward towards the ionization limit of the atom, because this time the higher the light frequency, the higher is the light intensity of the dynamical resonance /see Fig. 14/.

In the case of the curves ① and ② of Fig. 14 the $J_{Rj}/\bar{\nu}/$ functions cannot be approximated by the linear relationship $/I_{Rj} \sim \frac{1}{C}, \bar{\nu}/$. The reason for this effect and for the dependence of the Stark constant $/c'_j/$ on the level index /j/ will be explained in the following section.

10. DISCUSSION OF THE RESONANCE INTERACTIONS

To explain the observed behaviour of the resonance states in the light field it is necessary to refer to the term scheme of the He triplet system /Fig. 15/. First we notice that over and above the four-photon resonances of the n^3S, D, G states /fat arrows ① ② ③ /, the energy of which is shown in enlarged scale on the right-hand side of the figure,

in four-photon resonance is plotted for the different level widths /γ/ in Fig. 6.

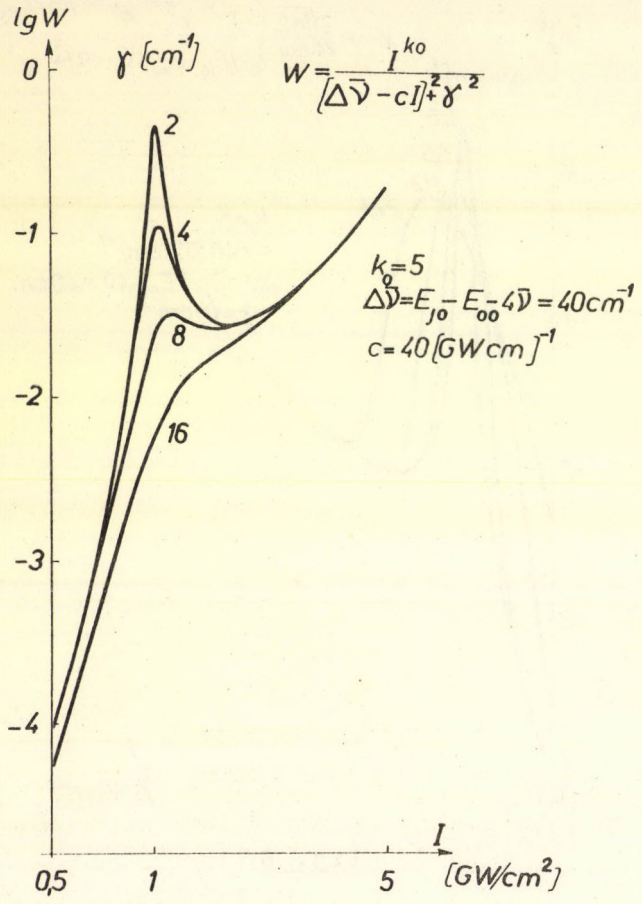

Fig. 6

In our measurement we used a laser working in multimode operation and therefore the laser could be considered as being a thermal light source. Agarwal [16] has shown that if the laser line shape is Lorentzian, so also will be the resultant dependence of the multiphoton ionization rate on the frequency, and the resultant level width $\gamma_r = N\gamma_L + \gamma$. Therefore, provided the assumption we have made are correct, the curves presented in Fig. 6 should describe the real situation with $\gamma = \gamma_r$. Given that the shape of the atomic level is always Gaussian, owing to Doppler effect, we obtain the curves shown in Fig. 7. These do not deviate strongly from the curves of Fig.6, because the Doppler level width /ΔE_D/ is always smaller than the laser linewidth /γ_L/.

The supposition of a Gaussian-shaped laser line /with linewidth $\Delta\bar{\nu}$/, on the other hand, yields curves of $W/I;\bar{\nu},\Delta\bar{\nu}$/ which deviate very considerably from the previous curves, as can be seen from Fig. 8. Furthermore, the supposition of Gaussian atomic level and Gaussian laser line, a case which was calculated by Rapoport et al. [17] in connection with the multiphoton absorption process, leads to almost identical curves.

The logW - logI function actually measured by us is presented in Fig. 9. The solid line was plotted on the assumption that the laser line is of Lorentzian shape. It is readily apparent that the experimental curve cannot be described by the supposition of a Gaussian laser line because of the large differences between it and the theoretical curve /cf. Fig. 8 and 9/.

Exact dynamical four-photon resonance occurs at the intensity $I_r = 6.10^8$ W/cm^2; that is, at this intensity, and neglecting the level

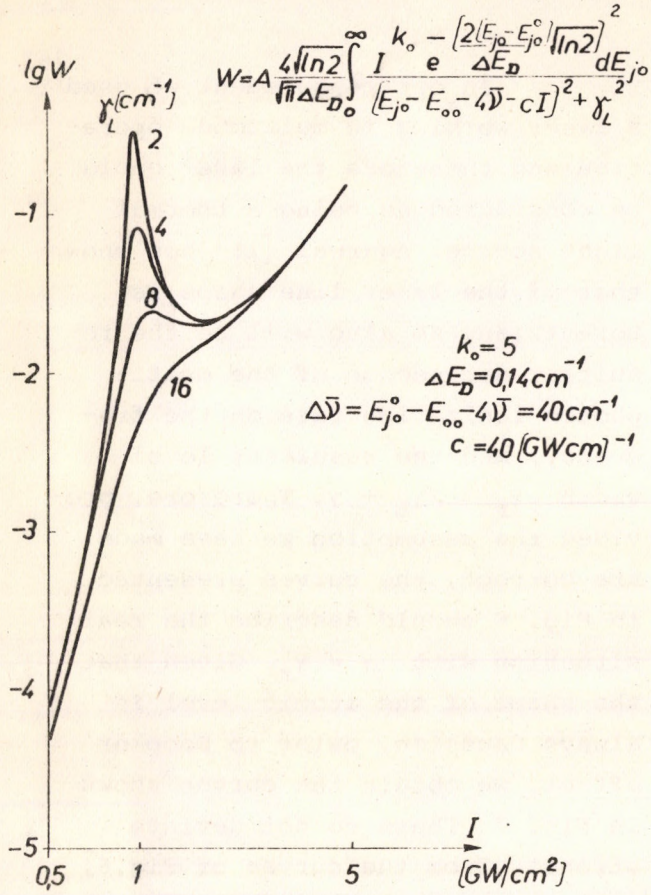

Fig. 7

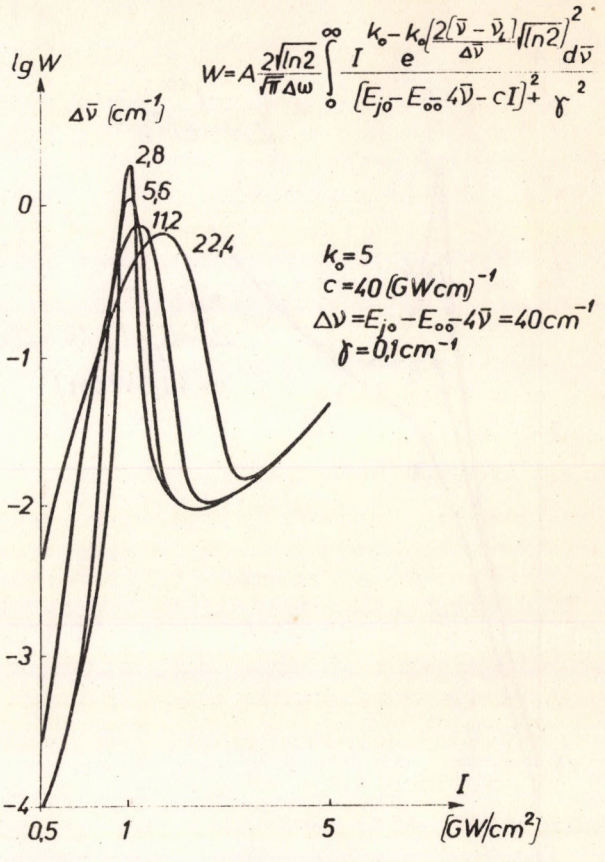

Fig. 8

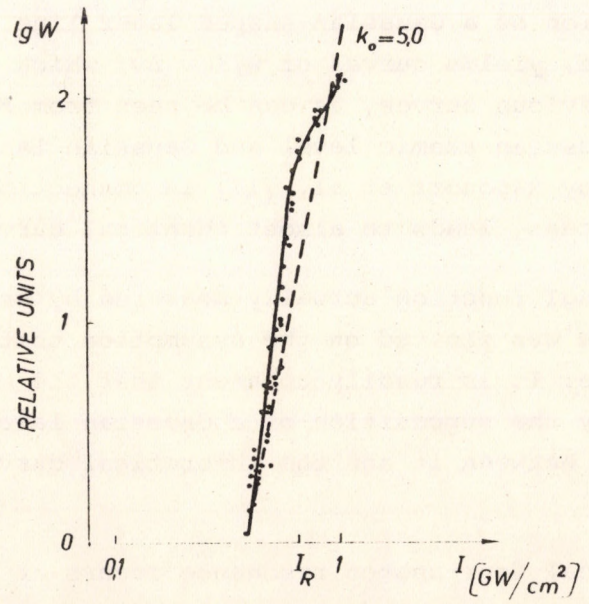

Fig. 9

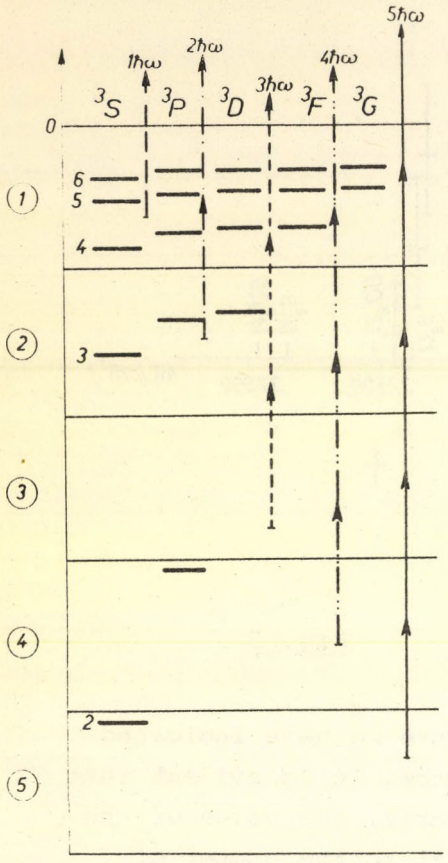

Fig. 2

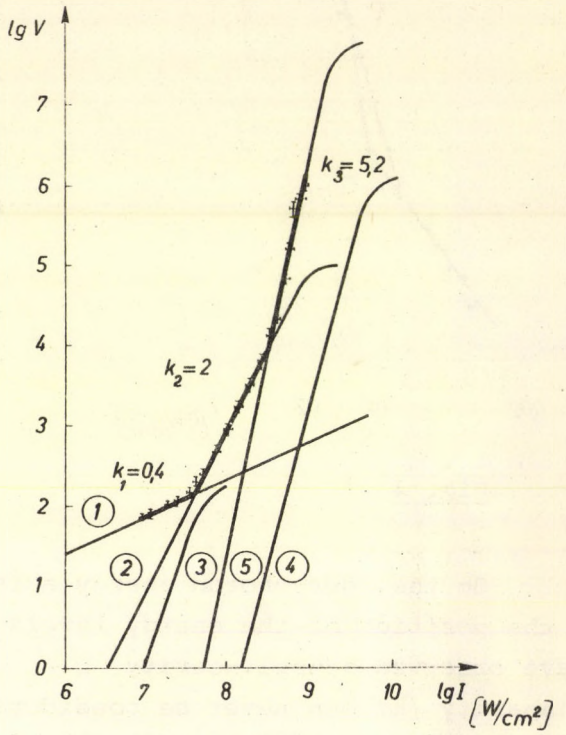

Fig. 3

section of the curve shows the contribution of two-photon ionization to the total ionization yield, while the upper section represents the five--photon ionization yield from the triplet metastable state.

5. NONLINEARITY

To investigate the influence of four-photon resonances on the five-photon ionization yield the frequency of the laser radiation was varied and the ionization yield versus light intensity function /$N_i \sim W/I;\bar{\nu}$/ was measured at different light frequencies [14]. Two typical curves are plotted in Fig. 4. It is apparent that, to a first approximation, the contribution of two-photon ionization is independent of the light frequency, whereas the five-photon ionization yield changes drastically. Adopting the usual linear approximation in the logW - logI coordinate system, we can plot the frequency dependence of the nonlinearity of the process /i.e. the value of the slope of this line /$k/\bar{\nu};I_1$/ /in logW - logI plot/ at constant light intensity /$I_1 = 2.10^8$ Wcm^{-2}/ /Fig. 5/.

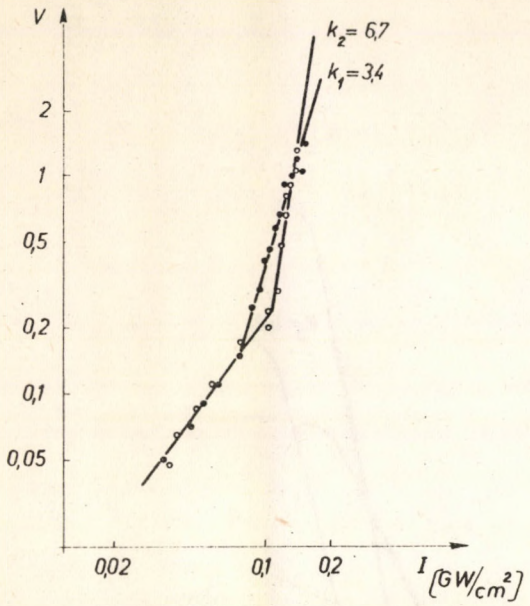

Fig. 4

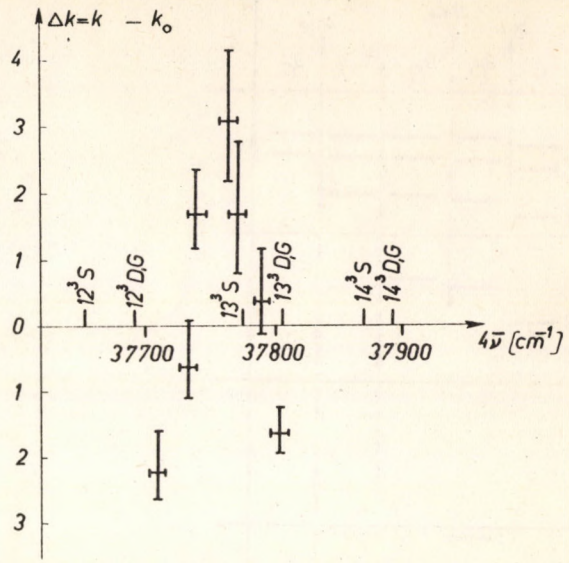

Fig. 5

On the four-photon energy axis of the figure we have indicated also the position of the energy levels of the He atom. It is evident that we have observed a nonlinearity $k > k_o$, so, clearly, the value of the nonlinearity /k/ can never be considered to be equal to the number of quanta absorbed during the ionization; this variation of the factor k with light intensity is a feature of the approximation of higher than k_o order, i.e. the consequence of a shift of levels in the high-intensity laser field. We should further note the fast change of the nonlinearity with frequency, which points to the existence of distinct atomic states even in the energy range close to the ionization limit.

6. GENERAL RATE VS. INTENSITY FUNCTION - STATISTICS - LINE SHAPE

During subsequent measurements it became obvious that the linear approximation in the logW - logI coordinate system is simply inadequate for describing the multiphoton ionization rate-light intensity dependence in the resonance case and over a wide range of light intensities [15]. We postulate that the required energy correction term $/\Delta E_j/I//$ to the four--photon resonance state /j/ depends linearly on the light intensity:

$$\Delta E_j(I) = C_j I \qquad /10/$$

The dependence of the five-photon ionization rate on light intensity $W/I;\bar{\nu},\gamma/$ resulting from expression /3/ on the assumption that the level is

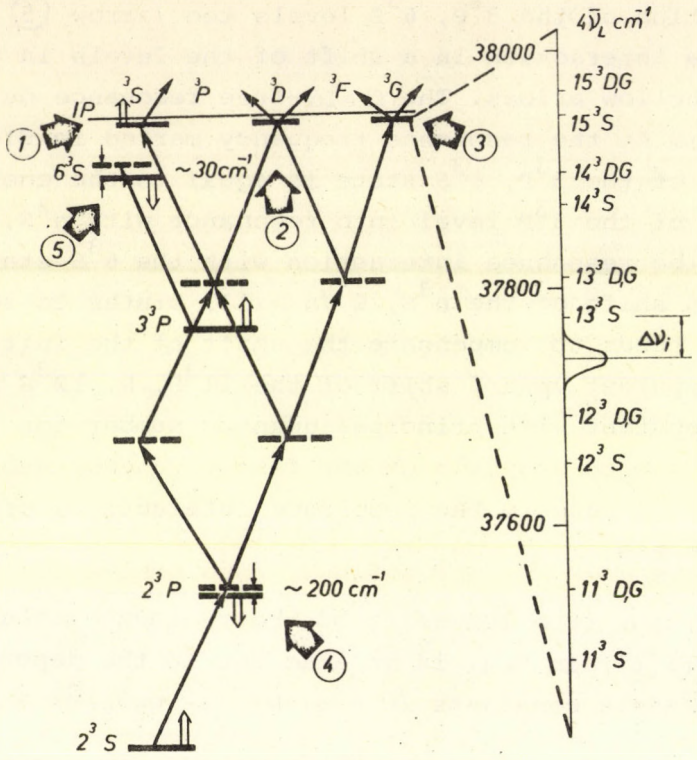

Fig. 15

there is another one-photon resonance of the 2^3S, 2^3P states /arrow ④/. The field-free resonance detuning is about 200 cm^{-1} and this resonance leads to a shift of the initial 2^3S state in the direction indicated by the hollow arrow /$\Longrightarrow$/. The theoretical value of the Stark shift of this level, as calculated on the basis of the Slater-type wave function, the experimental value of the term energies and the LS coupling scheme, was found to be ∿ 230 /GWcm/$^{-1}$.

The experimental value of the resultant Stark shift of the 2^3S, n^3S, D, G level /$\Delta E_o/I/ - \Delta E_j/I/ = c'_j I/$ was calculated from the tangent of the line of the resonance intensity function $I_{Rj}/4\bar{\nu}/$ to be ∿ 140 /GWcm/$^{-1}$ for n > 13. Taking into account that the four-photon resonance levels are shifted in the same direction as the initial level, due to the interaction with the continuum, the resultant Stark shift of the levels is excepted to be smaller than the shift of the initial level. Therefore the measured value /∿140 /GWcm/$^{-1}$/ may be considered to be in good agreement with the theoretical value / 230 /GWcm/$^{-1}$/.

To explain the measured dependence of the Stark constant on the level index /i.e. on the principal quantum number of the level/, it should be noted that over the above mentioned resonances there is the one-photon

resonance interaction of the 3^3P, 6^3S levels too /arrow ⑤ in Fig. 15/. The result of this interaction in a shift of the levels in the direction indicated by the hollow arroes. The field-free resonance detuning is very small / $\sim$ 30 cm^{-1}/. At the resonance frequency marked in Fig. 14 the energy difference of the 3^3P, 6^3S state is equal to the energy of the light quanta. The shift of the 3^3P level into resonance with n^3S, D /n > 11/ states caused by the resonance interaction with the 6^3S state, however, leads to an upward shift of the n^3S, D /n > 11/ states to the ionization limit. This shift tends to compensate the shift of the initial 2^3S level, resulting in an apparent upward shift of the 11^3S, D, 12^3S states and the change of Stark constant with principal quantum number for the $13,14^3S$, D states. This is the consequence of the fast frequency dependence of the Stark constant /$c_j/\bar{\nu}$// due to the resonance interactions of the 6^3S, 3^3P, n^3S, D /n > 11/ levels.

The deviation from linearity of the resonance intensity function $I_R/4\bar{\nu}$/ /curves ① ②/ in Fig. 14 is also due to the dependence of the various resonance Stark constants /c n^3S, D/$\bar{\nu}$/, $c_2{}^3S/\bar{\nu}$/ / on the frequency

The levels to which curves ① ③ of Fig. 14 refer cannot be identified because of the rapid variation of the Stark shift with frequency and the ensuing difficulty connected with extrapolation to zero field. The curves probably describe the dependence of the energy of the states with the principal quantum number 11 on the light intensity, but, as was mentioned before, 11^3S, D, 3^3P and 6^3S states are strongly coupled owing to the small field-free resonance detuning of the 3^3P, 6^3S states. In order to describe the motion of this system we would have to find the exact solution of the coupled differential equation of the motion of this states. Perturbation theory therefore does not provide an adequate answer in this case.

It should furthermore be noted that the Keldish-Kovarski parameter $\gamma = \frac{erE}{h\omega}$ [20, 21, 22] is greater than unity for the 11^3S, 11^3P states and therefore the perturbation method is not sufficient to describe the motion of this system either. The number of strongly coupled levels is increased and to describe the system we have to solve the coupled differential equation of motion of the 6^3S, 3^3P, 11^3S, P, D levels. Consequently, we cannot identify which 11^3S, 11^3D or 11^3P levels are involved.

Finally, it may be remarked that although resonance interaction of levels 6^3S, 3^3P plays no direct role in the multiphoton ionization it nevertheless leads to a significant disturbance of the system.

11. SUMMARY

To sum up: we have succeeded in investigating the distortion of the atomic helium system under the influence of a strong laser light and have found the shift of the atomic states lying in the energy range close to the ionization limit of the atom by measuring the dependence of the resonance multiphoton ionization rate on different parameters. Finally, we have experimentally determined the limit of application of the perturbation theory.

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