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Resonant multiphoton ionization of neon. Experimental determination of the polarizability of the $3p'[1/2]_1$ level

G. Baravian, J. Godart, and G. Sultan

Laboratoire de Physique des Plasmas, Université de Paris XI, Centre d'Orsay, 91405 Orsay Cedex, France*

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Multiphoton ionization of neon in the case of a 16-photon resonance with the $3p'[1/2]_1$ intermediate level is studied using a neodymium-glass laser. Experiments show that when the electric field E ranges from 2.2×10^7 to 3.5×10^7 V/cm, the number of collected charges takes a maximum value at 2.9×10^7 V/cm. A theoretical study on the basis of the formalisms due to Keldysh and Zon *et al.* predicts that, when an intermediate resonant level is considered, this maximum can be observed by the use of a laser beam of sufficiently large spectral width. This is the case in our experiment. The number of ionization events decreases when the resonance is destroyed, i.e., when the Stark shift of the level becomes larger than the spectral width of the laser, which, at 16 photons, is of the order of 100 Å. Good agreement between experiment and theory allows us to determine the polarizability of the resonant $3p'[1/2]_1$ level of neon which is found to be 21.8 Å^3. We have also determined an experimental cross section of multiphoton ionization of neon at $\lambda = 1.06$ μm.

I. INTRODUCTION

The problem of the multiphoton ionization (MPI) of a gas subject to the action of an intense coherent radiation field has been well investigated theoretically and experimentally in the last decade.[1] In our previous work[2] we reported the experimental verification of the predicted resonant behavior of the MPI probability by varying the incident laser wavelength at constant incident power. In that work the spectral width of the radiation was negligible.

In the present work, using a laser beam of large spectral width, another approach to the resonant aspect of MPI is taken: the resonance is destroyed if the electric field induced by the laser flux is strong enough to give rise to a large Stark shift of the intermediate resonant level.

In a previous Letter[3] we reported the results of a MPI experiment in neon using a neodymium-glass laser. We showed that a plot of the number of ionization events versus the incident laser flux passes through a maximum for a certain value of the flux and then decreases. Taking into account the influence of the laser intensity, the processes leading to this result can be described as follows: At low flux, in the case of exact resonance between the additive energy of 16 photons ($9440\times16=151\,040$ cm^{-1}) and the energy of the $3p'[\frac{1}{2}]_1$ neon atomic level (151 040 cm^{-1}), 16 laser photons simultaneously interact with a neon atom. This excitation by 16 photons is possible since the $3p'[\frac{1}{2}]_1$ level has the same parity as the fundamental level and an even number of photons conserves the parity for the transition. The excitation of this level is the first stage of the ionization process. The second stage, representing the ionization of the resonant level, takes place by absorption of three photons. This does not mean that we have a "two-step" process: the probabilities of these two steps are not independent.[4] The probability of this second process is much larger than that of the first. If there were no resonant level, the ionization would be a process with 19 photons interacting simultaneously, and the probability for such a process is lower than the probability with an intermediate resonant level.

Because of the spectral width of the incident radiation, there is an overlap of the laser with the resonant atomic level. The experimental intensity profile of the laser is Gaussian with full width at half-maximum (FWHM) Γ of 25 Å (2.76×10^{-3} eV). This corresponds to a spectral extent of $\Gamma_L=\sqrt{16}\,\Gamma=1.1\times10^{-2}$ eV in the case of the interaction with $N=16$ photons.

When the laser flux is increased, the Stark shift of the intermediate level leads to a detuning of the resonance. The overlap of the laser with the atomic level may be decreased, and for certain values of flux one finds a change in the slope of the curve of $\log N_i$ vs $\log\phi$, where N_i is the ion yield and ϕ is the laser flux. This change in slope is due to a decrease in the ionization probability.

Figure 1 represents the evolution of the resonant $3p'[\frac{1}{2}]_1$ level and of the neighboring levels, with increasing flux of the spectrally broad neodymium laser radiation. As seen in Fig. 1(a), when the laser flux is low, there is no shift or broadening, and thus the interaction is resonant. Figure 1(b) shows that for higher laser flux, the $3p'[\frac{1}{2}]_1$ level is slightly shifted and the effective flux which is on resonance is decreased. Figure 1(c) shows that for extremely strong values of the laser flux, the $3p'[\frac{1}{2}]_1$ level is broadened and shifted completely out of resonance with the 16 laser photons.

This qualitative explanation assumes that neigh-

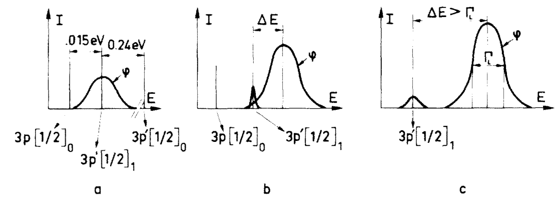

FIG. 1. Qualitative behavior of the resonant $3p'[\frac{1}{2}]_1$ level for a growing light flux. (a) Low flux, good resonance between ϕ and the $3p'[\frac{1}{2}]_1$ level. (b) Higher flux; the effective part of the flux which is in resonance decreases. (c) Extremely high flux; the resonance is destroyed by the shift ΔE of the $3p'[\frac{1}{2}]_1$ level. ϕ represents the 16-photon spectral distribution of the light flux.

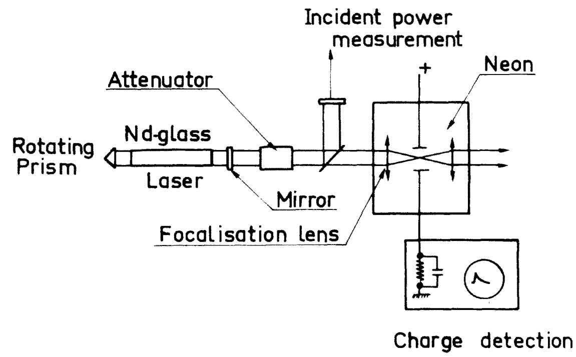

FIG. 2. Experimental setup.

boring levels are shifted so that the energy gap between these levels and the spectral position of the laser radiation is increased.

The paper is organized as follows: In Sec. II we describe the experimental setup and give a curve $\log N_i$ vs $\log E$, where N_i is the number of charges created by MPI under the action of the electric field E associated with the laser radiation. In Sec. III we give a theoretical expression for the MPI probability (Sec. III C) which takes into account the width Γ of laser radiation, the broadening Γ_s, and the Stark shift $\Delta E = \frac{1}{2}\alpha_s E^2$ of the resonant level, where α_s is the polarizability of the resonant level. A first derivation shows that the ionization probability of the resonant level is very high (Sec. III A) and that the direct ionization of a neon atom by a 19-photon process is not accessible in the case of our experimental parameters (Sec. III B). In Sec. IV we determine α_s. A theoretical expression for the apparent slope T of the curve $\log N_i$ vs $\log\phi$ (ϕ is the laser flux) is found as a function of α_s, Γ, and E (Sec. IV A). From this relation we can determine the polarizability α_s (Sec. IV B). With the aid of α_s thus determined, we calculate a value of T which is in good agreement with the experimental values (Sec. IV C). Finally, in Sec. V we give a numerical value for the probability of MPI.

II. EXPERIMENTAL SETUP AND RESULTS

Figure 2 shows the usual experimental setup. The light emitted by a neodymium-glass laser—$\lambda = 1.06$ μm, pulse duration time of 20 nsec, and energy of 3 J—is focused with a lens of $f = 4$ cm focal length into a chamber containing neon gas at a pressure of 10^{-3} Torr. The measured divergence of the laser beam is $\theta = 1.25\times 10^{-3}$ rad. During the experiment the chamber is traversed by a flow of neon. Before the experiment, the chamber is pumped to a pressure of 10^{-7} Torr in order to avoid ionization of impurities. A 100-V/cm field is applied between two electrodes placed at each side of the focal region to collect the charges produced. These electrodes are partly shielded in order to avoid parasitic detection. To avoid collisions, the pressure is chosen so that the mean free path is much larger than the distance between the electrodes, but this pressure must not be too low, since the number of charges produced is proportional to the pressure. The detection system gives an oscilloscope signal whose leading-edge amplitude is proportional to the number of ions collected. The incident radiation intensity is varied by means of neutral-density filters placed in the beam. The light beam is linearly polarized. The optical electrical field in the interaction region varies from 2×10^7 to 4×10^7 V/cm. Figure 3(a) gives typical examples of oscillograms for three increasing values of electric field, and Fig. 3(b) gives the curve N_i as a function of the electric

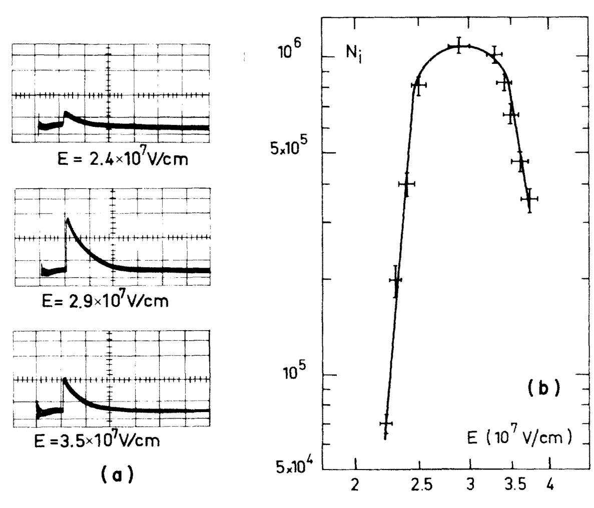

FIG. 3. (a) Typical oscillograms of signal detected for three increasing electric fields. Vertical sensitivity is 5 mV/division. Horizontal sensitivity is 500 μsec/division (1 vertical division $\simeq 6\times 10^5$ ions). a_1: $E = 2.4\times 10^7$ V/cm; a_2: $E = 2.9\times 10^7$ V/cm; a_3: $E = 3.5\times 10^7$ V/cm. (b) Number of charges detected vs the laser electric field.

field, deduced from the oscillogram measurements. We see that a maximum value of the number of the collected charges is reached for an electric field of 2.9×10^7 V/cm. At the beginning, for lower laser fluxes, the relation between N_i and ϕ is $N_i \propto \phi^{12}$; we have never found the relation $N_i \propto \phi^{16}$ as we had expected. We notice that this result agrees well with the value obtained by Agostini *et al.*,[5] who, under similar conditions, give a slope $T = 13\pm1$ for an electric field of $E = 2.3\times10^7$ V/cm.

Let us remark that the experiments carried out by Delone *et al.*[6] on the MPI of potassium (whose ionization needs a four-photon interaction for a Nd laser radiation) lead, in the case of no resonance, to a slope equal to 4 at low flux and tending to zero when the flux is increased. In the case of three-photon resonant interaction, they have found a slope decreasing from 1.5 to zero. Similar experiments by Alimov *et al.*[7] on xenon show that the slope varies from 14 to 2.5 when the flux is increased. In any case, the experiments with high laser flux always lead to values of slope smaller than the value obtained by the relation $[\![1 + E_i/\hbar\omega]\!]$, where E_i is the ionization energy of an atom, $\hbar\omega$ is the energy of the photon, and $[\![\ ]\!]$ stands for the integer part. When the slope becomes zero most of the workers ascribe it to sample depletion, which is often called saturation.

All of our experimental results cannot be explained, however, by the mechanism of sample depletion, as is widely believed. This is because there exists a region of negative slope on the curve of $\log N_i$ vs $\log\phi$ and because the number of collected ions is smaller than the number N_0 of atoms in a sphere of radius $f\theta$ ($N_0 \simeq 1.8\times10^7$ compared to the number collected $\simeq 10^6$).

III. PROBABILITY OF MULTIPHOTON IONIZATION

The MPI probability is usually given, in the case of an intermediate resonant level, by

$$W_0^c = W_0^s W_s^c , \tag{1}$$

where W_0^s represents the probability of excitation of the resonant level s from the ground state and W_s^c the probability of ionization of the level s. In the case considered these probabilities concern 16- and 3-photon interactions respectively.

A. Calculation of W_s^c

The expression for the ionization probability of an excited level has been given by Keldysh[8] in a semiclassical formalism as

$$W_s^c \simeq \omega \left(\frac{E_i - E_s}{\hbar\omega}\right)^{3/2} \exp\left[2[\![1 + (E_i - E_s)/\hbar\omega]\!] - \frac{E_i - E_s}{\hbar\omega}\left(1 + \frac{1}{\gamma^2}\right)\right]\left(\frac{1}{4\gamma^2}\right)^{[\![1+(E_i-E_s)/\hbar\omega]\!]}$$
$$\times\Phi([2[\![1 + (E_i - E_s)/\hbar\omega]\!] - 2(E_i - E_s)/\hbar\omega]^{1/2}), \tag{2}$$

where ω is the angular frequency of laser radiation. For the Nd laser we have $\omega = 1.78\times10^{15}$ sec^{-1}; E_i is the ionization energy (21.559 eV), E_s the energy of the $3p'[\frac{1}{2}]_1$ level of neon, $E_i - E_s = 2.837$ eV, Φ is the error function, and

$$[\![1 + (E_i - E_s)/\hbar\omega]\!] = 3.$$

The expression (2) is valid for $\gamma > 1$, where $\gamma = \omega/\omega_t$, with

$$\omega_t = eE/[2m(E_i - E_s)]^{1/2} ;$$

E stands for the intensity of the electric field associated with the laser radiation. In our experiment, we find $\omega_t(\text{sec}^{-1}) = 1.76\times10^7 E$ (V/cm). For a variation of the electric field from 10^7 to 5×10^7 V/cm the extrema for ω_t become 1.76×10^{14} and 8.80×10^{14} sec^{-1}, respectively. Under these conditions $\gamma \simeq 10^8/E$ (V/cm) varies from 10 to 2. With $\Phi(1.07) = 0.87$ in (2) we obtain

$$W_s^c = 3.26\times10^{15}\gamma^{-6}e^{-2.425/\gamma^2}$$

or

$$W_s^c = 3.05\times10^{-33}E^6 e^{(-2.37\times10^{-16}E^2)},$$

with W_s^c in sec^{-1} and E in V/cm. Thus for $10^7 < E(\text{V/cm}) < 5\times10^7$, we have $2.98\times10^9 < W_s^c(\text{sec}^{-1}) < 2.63\times10^{13}$. We note that the probability W_s^c varies with E as expected, i.e., $W_s^c \propto \gamma^{-6} \propto E^6 \propto \phi^3$ (three-photon interaction). We see also that for the lowest electric field the complete ionization of the s level is achieved in a time of the order of 0.3 nsec, which can be considered as very short compared to the time of the laser pulse duration (20 nsec). In the following we therefore consider only the excitation probability W_0^s, in the case where ionization is through a resonant level.

B. Calculation of W_0^c

If we do not take the resonant level into account, we can calculate a probability W_0^c of the direct MPI by use of expression (2), in which the energy $E_i - E_s$ is replaced by the ionization energy of the fundamental state (21.559 eV). The result is

$$W_0^c = 1.715\times10^{-307}E^{38}e^{(-2.37E^2 10^{-16})}, \tag{3}$$

with W_0^c in sec^{-1} and E in V/cm, which shows the usual behavior in ϕ^{19} ($\phi \propto E^2$) for W_0^c.

By using the previous values of the electric field, we obtain

$$1.675\times10^{-41} \leq W_0^c(\sec^{-1}) \leq 3.5\times10^{-15} .$$

The number of ions produced by this process is $dn = W_0^c n_0 v_0 \tau$, where n_0 is the density of neutral atoms, v_0 is the interaction volume, and τ is the duration of the laser pulse. For $\tau = 20$ nsec and experimental values of v_0 and n_0, dn is much smaller than unity and no ionization occurs.

We note that for the electric field considered, there is no possibility of ionization of the neon atom by tunnel effect, since the criterion for this process is $\gamma \leq 1$, where

$$\gamma = \gamma_0 = \frac{\omega}{(\omega_t)_0} = \frac{\omega(2mE_i)^{1/2}}{eE} \simeq \frac{2.8\times10^8}{E(\text{V/cm})} .$$

For E varying between 10^7 and 5×10^7 V/cm this gives $5.6 < \gamma_0 < 28$.

C. Expression of W_0^s

W_0^s is a function of the laser linewidth Γ, the level width γ_s, the broadening W_s^c, and the shift ΔE caused by the radiation field. The formalisms used by Keldysh[8] and Bebb and Gold[1] cannot be directly applied to our experiment, since they have used a very narrow laser spectrum. In the case where the laser spectrum is Gaussian with half-width Γ, and where the s level has a Gaussian profile with half-width Γ_s, we can use the formula given by Zon *et al.*,[9]

$$W_0^s = K_1 \frac{\phi^N}{(\Gamma_s^2 + N\Gamma^2)^{1/2}} \exp\left(- \frac{(E_s - N\hbar\omega + \Delta E)^2}{\Gamma_s^2 + N\Gamma^2} \right), \tag{4}$$

where the constant K_1 is a function of ω and of atomic parameters. In (4) $N = 16$ and $\Gamma_s = W_s^c + \gamma_s \simeq W_s^c$. Let us recall that for the neon atom ΔE is not the linear but the quadratic Stark effect. The linear effect occurs only for hydrogen where the levels are degenerate,[10] or for other atoms whose levels are very close to the continuum and can thus be treated as degenerate. We note that as previously mentioned in Sec. I, the expression of W_0^s is not independent of W_s^c because, for example, the width Γ_s due to the ionization affects the quantity W_0^s.

To be precise, we are now in a position to compare Eq. (4) with the results obtained by Keldysh. If we assume that $E_s - N\hbar\omega = 0$ and that the exponent is much smaller than unity, we obtain

$$W_0^s = K_1 \phi^N (\Gamma_s^2 + N\Gamma^2)^{1/2} / (\Delta E^2 + \Gamma_s^2 + N\Gamma^2); \tag{5}$$

when N is an even number and the quadratic Stark effect is taken into account, Keldysh's formula is written

$$W_0^s = \frac{1}{4}\left| J_{N/2+1}\left(\frac{e^2E^2\alpha_s}{4\hbar\omega}\right) + J_{N/2-1}\left(\frac{e^2E^2\alpha_s}{4\hbar\omega}\right)\right|^2 \times \frac{|V_{0s}^{(1)}|^2\Gamma_s}{\Delta E^2 + \Gamma_s^2} . \tag{6}$$

Although the complete formulation involves a summation over all atomic levels, it is easy to show that in the presence of a resonant level this single term dominates the sum. In Eq. (6), $|V_{0s}^{(1)}|$ is the transition matrix element from the ground state to the s state. The term $V_{0s}^{(1)}$ is the correction to $V_{0s} = \int \psi^* E \psi_0 dv$ in the first-order perturbation theory. $J_\nu(z)$ is the Bessel function of order ν. The expression (6) can be rewritten

$$W_0^s = \frac{1}{4}\left| \frac{N}{(e^2E^2\alpha_s)/4\hbar\omega} J_{N/2}\left(\frac{e^2E^2\alpha_s}{4\hbar\omega}\right)\right|^2 \frac{|V_{0s}^{(1)}|^2\Gamma_s}{\Delta E^2 + \Gamma_s^2} . \tag{7}$$

The term $e^2E^2\alpha_s$ represents the Stark shift, which is always very small compared to $\hbar\omega$. We can write Eq. (7) in the simplified form

$$W_0^s = K_2 \phi^N \Gamma_s / (\Delta E^2 + \Gamma_s^2) , \tag{8}$$

where K_2 is constant. The expression (8) has the same form as (5), in which Γ is taken to be equal to zero.

IV. POLARIZABILITY α_s OF THE RESONANT LEVEL

In this section we wish to determine the polarizability α_s, by using the theoretical expression of the probability W_0^s and our experimental results.

A. Theoretical expression of the apparent slope T

To evaluate W_0^s numerically, Eq. (4) is of no practical use, because the constant K_1 is unknown. For weak electric fields, the Stark shift ΔE is small and thus W_0^s varies as ϕ^{16}. This behavior is quite different for strong electric fields where Eq. (4), $W_0^s = f(\phi)\phi^N$, can be put into the form $W_0^s = A\phi^T$, T being an apparent slope different from N and A being a constant. Then, using the relation $T(\phi) = d\ln W_0^s / d(\ln\phi)$ we obtain from Eq. (4)

$$T = N - 2\frac{(\Delta E)^2}{\Gamma_s^2 + N\Gamma^2}\left(1 - \frac{3\Gamma_s^2}{\Gamma_s^2 + N\Gamma^2}\right) - \frac{3\Gamma_s^2}{\Gamma_s^2 + N\Gamma^2} , \tag{9}$$

in which Γ_s is proportional to ϕ^3 and ΔE proportional to ϕ. Values of the slope T obtained by this expression can be compared with the value obtained experimentally [Fig. 3(b)]. We have already seen that when electric fields become stronger Eq. (4) will be invalid above a certain threshold. Moreover, this equation renders account of the experiment only as long as the apparent slope T is smaller than N. This leads to the condition

$$1 - 3\Gamma_s^2/(\Gamma_s^2 + NT^2) > 0,$$

which gives an upper limit for the values of the width Γ_s and of the electric field E. From the above-mentioned inequality we obtain $\Gamma_s < 0.707\Gamma_L = 7.8\times10^{-3}$ eV and $E < 4.3\times10^{7}$ V/cm.

B. Value of the polarizability

To calculate T we must know ΔE and consequently the polarizability α_s. The formal expression for α_s is[11]

$$\alpha_s(\omega) = \sum_m |\langle s|z|m\rangle|^2 \left(\frac{1}{\omega_{sm}+\omega} + \frac{1}{\omega_{sm}-\omega}\right), \tag{10}$$

where ω is the angular frequency of the incident radiation and ω_{sm} is the difference in energy between the level s and a level m in the case where the transition $s \longleftrightarrow m$ is optically allowed.

Obviously the dominant terms in the sum in Eq. (10) are those for which $\omega_{sm} \simeq \omega$. The factor $|\langle s|z|m\rangle|^2$ depends on the statistical weight and oscillator strength, and the values for the transition $s \rightarrow m$ corresponding to 6599 and 6163 Å are given in Ref. 12. For these transitions we find that ω_{sm}, the corresponding energy of which is about 1.9 eV, is not very far from the energy of the photons of the laser beam (1.17 eV). A value of the polarizability α_s obtained from these two lines takes a value of 10^{-22} cm^3. A diagram (Fig. 4) of energy levels of the neon atom shows that we have to take into account all transitions $2p^5 4s$ and $2p^5 3d$ to the level $3p'[\frac{1}{2}]_1$ for which $\omega_{sm} \simeq \omega$. The contribution to α_s from the transitions with $\omega_{sm} < \omega (2p^5 4s)$ and those with $\omega_{sm} > \omega$ $(2p^5 3d)$ give values similar but of opposite sign. However, the

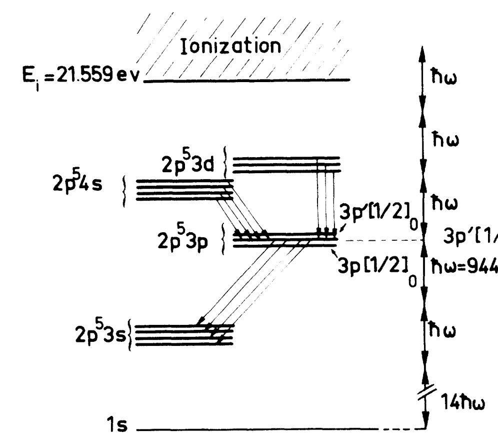

FIG. 4. Simplified energy-level diagram for the neon atom.

differences in numerical values of the matrix elements $|\langle s|z|m\rangle|^2$ calculated by theoreticians[13] do not allow us to determine α_s correctly by this approach.

To circumvent this drawback, we have tried to determine α_s by using the experimental slope T. We assume that in the expression $N_i = W_0^s v_0 n_0 \tau$ only the factor W_0^s depends on ϕ, so that $N_i \propto \phi^T$.

Experimentally we find $T=0$ for $E = 2.9\times10^7$ V/cm. In Eq. (9) we put $T=0$. By using

$$\Delta E = 1.73\times10^6 \alpha_s E^2,$$

with ΔE in eV, α_s in cm^3, and E in V/cm, we find $\alpha_s = 2.18\times10^{-23}$ cm^3. The region near $T=0$ being flat ($T \simeq 0$ for $E = 2.9\pm0.1\times10^7$ V/cm), we find $\alpha_s = 2.18\pm0.14\times10^{-23}$ cm^3. This value of α_s is comparable to that obtained for the level $11p$ of neon irradiated by a ruby laser ($\alpha_{11p} = 5.7\times10^{-24}$ cm^3).[2]

C. Dependence of the apparent slope on the electric field

Using the value $\alpha_s = 2.18\times10^{-23}$ cm^3 previously determined, we give in Table I numerical results on the shift ΔE, the broadening Γ_s, and the apparent slope T as a function of the electric field E. The relations

$$\Delta E = 3.77\times10^{-17} E^2,$$

$$\Gamma_s = \hbar W_s^c = 2.006\times10^{-48} E^6 e^{-2.37\times10^{-16}E^2},$$

where ΔE and Γ_s are in eV and E is in V/cm, have been used.

The value of α_s has been determined by using a point on the experimental curve of $\log N_i$ vs $\log E$ [Fig. 3(b)], i.e., the point E where $T=0$. In order to calculate T we have reintroduced this value of α_s in Eq. (9). A comparison between the numerical results of T given in Table I with our experimental results deduced from Fig. 3(b) for E varying from 2.2×10^7 to 3.5×10^7 V/cm shows a reasonably good agreement. This agreement allows us to claim that this method of determining α_s introduces no serious error.

We see that when we take into account the above-obtained values of ΔE of the $3p'[\frac{1}{2}]_1$ level, the neighboring level $3p'[\frac{1}{2}]_0$ on the higher-energy

TABLE I. Values of the shift ΔE, the broadening Γ_s, and T for different values of the electric field E (V/cm).

E	ΔE (eV)	Γ_s (eV)	T
10^7	3.77×10^{-3}	1.96×10^{-6}	15.77
2×10^7	1.51×10^{-2}	1.17×10^{-4}	12.27
2.5×10^7	2.36×10^{-2}	4.22×10^{-4}	6.94
2.9×10^7	3.17×10^{-2}	9.77×10^{-4}	0
3×10^7	3.39×10^{-2}	1.18×10^{-3}	−2.06
3.5×10^7	4.62×10^{-2}	2.76×10^{-3}	−11.3

side, which is 0.24 eV distant, has no effect on the process leading to the detuning of the resonance. The $3p[\frac{1}{2}]_0$ level, on the lower-energy side, has, however, an energy separation of 0.015 eV with respect to the resonant level. This energy is comparable to the shift calculated for an electric field of about 2×10^7 V/cm. Considering the fact that our experiments show a negative slope for these values of the electric field, we think that the shift of the $3p[\frac{1}{2}]_0$ level tends to increase the separation between this and the resonant levels. Otherwise the $3p[\frac{1}{2}]_0$ level should occupy the place of the $3p'[\frac{1}{2}]_1$ level and we would have never found a detuned situation for the resonance.

V. IONIZATION PROBABILITY

The ionization probability can be experimentally determined by assuming that the interaction volume is constant. By use of the relation $N_i = W_0^s v_0 n_0 \tau$, where $n_0 = 3.54\times10^{13}$ cm^{-3}, $\tau = 2\times10^{-8}$ sec, and $v_0 = 5.24\times10^{-7}$ cm^3, we find $W_0^s = 2.7N_i$. Thus W_0^s varies from 10^5 to 3×10^6 sec^{-1}. The lowest value is equal to that given by other authors.[5]

With our experimental results, we can determine by virtue of Eq. (4) a generalized cross section for ionization, usually expressed in ϕ^{-N} sec^{-1}. When we take in Eq. (4)

$$\chi = K_1/(\Gamma_2^s + N\Gamma^2)^{1/2} \simeq K_1/4\Gamma,$$

with $N\Gamma^2 > \Gamma_s^2$, we then find for the ionization probability

$$\chi = (5\pm3.5)\times10^{-183}\,\mathrm{sec}^{-1}\,(\mathrm{W\,cm}^{-2})^{-16}$$
$$= (1.16\pm0.8)\times10^{-482}\,\mathrm{sec}^{-1}\,(\mathrm{photons\,cm}^{-2}\,\mathrm{sec}^{-1})^{-16}\,.$$

VI. CONCLUSION

To summarize, our findings are as follows:

(i) The direct multiphoton ionization of neon by 1.06-μm radiation is a process involving 19 photons; its small probability renders it unobservable at available values of laser flux. In fact, the ionization process takes place through an intermediate resonant level when moderate flux values are used. This process is essentially governed by the excitation probability W_0^s with 16 photons of the s ($3p'[\frac{1}{2}]_1$) level, in which is included the three-photon ionization probability W_s^c of the excited level. However, the ionization process is not a two-step process, because there is a definite correlation between the two transitions (excitation and ionization of the s level).[4]

(ii) The probability W_0^s can be determined as a function of the radiation flux by using the formalism of Zon *et al.*,[9] which gives

$$W_0^s = K(\phi)\phi^{T(\phi)}\ ,$$

where $K(\phi)$ and $T(\phi)$ are functions of the flux ϕ. In this expression, the broadening of the s level, its shift under the influence of the electric field of the radiation as well as the width of the laser radiation, are included. Therefore the slope

$$T = d(\ln W_0^s)/d(\ln\phi)$$

turns out to be a function of those parameters. For weak electric fields, $E \leq 10^7$ V/cm, the slope is equal to 16 and is less than 16 for $E > 10^7$ V/cm. When the electric field increases above the value 10^7 V/cm, the slope T decreases, passes through zero, and then changes sign. This prediction is in good agreement with our experimental results, which show that the number of charges created by multiphoton ionization takes a maximum value at $E = 2.9\times10^7$ V/cm. This variation of the slope is explained by a shift of the s level due to the quadratic Stark effect.

(iii) An analysis of the experimental results allows us to determine the polarizability of the $3p'[\frac{1}{2}]_1$ level of neon, which is found to be 21.8 Å^3. The generalized cross section for multiphoton ionization does not depend markedly on the laser flux if $E \leq 3.5\times10^7$ V/cm. The proposed value is $\chi \simeq 10^{-482}$ cm^{32} sec^{15}.

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*Associated with the Centre National de la Recherche Scientifique.

[1]H. B. Bebb and A. Gold, Phys. Rev. 143, 1 (1966); 176, 420 (1968); G. S. Voronov and N. B. Delone, Zh. Eksp. Teor. Fiz. 50, 78 (1966) [Sov. Phys.-JETP 23, 54 (1966)]; H. R. Reiss, Phys. Rev. A 1, 803 (1970); M. Luvan, G. Mainfray, C. Manus, and I. Tugov, *ibid.* 7, 91 (1973); Y. Gonthier and M. Trahin, *ibid.* 7, 1899 (1973); C. B. Collins, B. W. Johnson, D. Popescu, G. Musa, M. L. Pascu, and I. Popescu, *ibid.* 8, 2197 (1973); C. Cohen-Tannoudji, J. Dupont-Roc, C. Fabre, and G. Grynberg, *ibid.* 8, 2747 (1973); D. T. Alimov, N. K. Berezhetskaya, G. A. Delone, and N. B. Delone, Zh. Eksp. Teor. Fiz. 64, 1178 (1973) [Sov. Phys.-JETP 37, 599 (1973)]; D. Popescu, C. B. Collins, B. W. Johnson, and I. Popescu, Phys. Rev. A 9, 1182 (1974); A. Decoster, *ibid.* 9, 1446 (1974); P. Lambropoulos, Phys. Rev. 168, 1418 (1968); Phys. Rev. Lett. 30, 413 (1973); B. A. Zon and B. G. Katsnelson, Zh. Eksp. Teor. Fiz. 65, 947 (1973) [Sov. Phys.-JETP 38, 470 (1974)]; J. S. Bakos, Adv. Electron. Electron Phys. 36, 57 (1974); L. Armstrong, Jr., B. L. Beers, and S. Feneuille, Phys. Rev. A 12, 1903 (1975); B. L. Beers and L. Armstrong, Jr., *ibid.* 12, 2447 (1975); C. G. Morgan, Rep. Prog. Phys. 38, 621 (1975); A. M. F. Lau, Phys. Rev. A 13, 139 (1976).

[2]G. Baravian, R. Benattar, J. Bretagne, J. L. Godart, and G. Sultan, Appl. Phys. Lett. 16, 162 (1970); R. Benattar and G. Sultan, J. Appl. Phys. 45, 539 (1974).
[3]G. Baravian, R. Benattar, and G. Sultan, Appl. Phys. Lett. 25, 291 (1974).
[4]P. Lambropoulos, Phys. Rev. A 9, 1992 (1974).
[5]P. Agostini, G. Barjot, J. F. Bonnal, G. Mainfray, C. Manus, and J. Morellec, IEEE J. Quantum Electron. QE-4, 667 (1968).
[6]G. A. Delone, N. B. Delone, and G. K. Piskova, Zh. Eksp. Teor. Fiz. 62, 1272 (1972) [Sov. Phys.-JETP 35, 672 (1972)].
[7]D. T. Alimov, N. B. Delone, B. A. Zon, and B. G. Katsnelson, FIAN Report No. 191 (1973) (unpublished).
[8]L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys.-JETP 20, 1307 (1965)].
[9]B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. 61, 968 (1971) [Sov. Phys.-JETP 34, 515 (1972)]; Dokl. Akad. Nauk SSSR 188, 560 (1970) [Sov. Phys.-Doklady 14, 904 (1970)]; Opt. Spektrosk. 38, 13 (1975) [Opt. Spectrosc. 38, 6 (1975)].
[10]A. M. Bonch-Bruevich and V. A. Khodovoi, Usp. Fiz. Nauk 93, 71 (1967) [Sov. Phys.-Usp. 10, 637 (1968)].
[11]V. A. Davydkin, B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. 60, 124 (1971) [Sov. Phys.-JETP 33, 70 (1971)].
[12]C. E. Moore, *Atomic Transition Probabilities, Hydrogen through Neon,* Publication No. NSRDS-NBS 4 (Natl. Bur. Stand., U.S. GPO, Washington, D. C., 1966), Vol. I.
[13]P. W. Murphy, J. Opt. Soc. Am. 58, 1200 (1968); A. V. Loginov and P. F. Gruzdev, Opt. Spectrosk. 37, 817 (1974) [Opt. Spectrosc. 37, 467 (1974)].