

## A new, non-local quantum electrodynamics : its effect on multiphoton single and double ionization\*

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**Abstract** . By considering a new, non-local quantum electrodynamics, we have shown that the non-sequential double ionization of atoms occur due to the phase correlated two-photon excitation of electrons. For this phase correlated excitation, two-photon rate depends linearly on laser intensity. However, the local nature of the radiation field gives rise to conventional multiphoton transitions, where the two-photon rate is proportional to the square of the laser intensity. We have shown that  $n$ -photon transitions in an experiment can be modeled as a series of these two types of transitions and hence several channels will run parallel to each other. The channels for  $n$ -photon transitions are (i) a series of phase correlated transitions giving rise to non-sequential double ionization, (ii) a series of conventional dipole transitions giving rise to sequential ionization and (iii) a mixture of phase correlated and phase non-correlated transitions. We have shown that, with the increase in laser intensity the channel comprising of only the phase correlated excitations becomes significant and gets saturated before the sequential channel can dominate, giving rise to a kink-like structure in the intensity variation of double ionization. This feature has been observed in the experiments on double ionization of He. We have also provided a physical basis for the damping of non-sequential double ionization at the smaller wavelength, which is also an experimentally observed feature.

**Keywords** . Multiphoton ionization, double ionization, non-local QED

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### 1. Introduction

During the last decade, the investigations in the field of double ionization of rare gas atoms have shown that the intensity dependence of doubly charged ions shows a kink, which can not be explained by the single rate of ionization. Two mechanisms [1] for double ionization have been proposed : (i) sequential and (ii) direct or non-sequential (NS) double ionization. In the sequential process, the atom is first singly ionized and then the ionization of singly charged ion occurs by absorbing more photons. In the direct process, independent electron model does not work and two electrons are considered to be ejected simultaneously to produce doubly charged ions. For this process, two mechanisms have been proposed [2,3] : (i) shake-off model and (ii) re-scattering model. But there are controversies over applicability of these two models;

some experimentalists [4] support re-scattering model and some others [5] support shake-off model. Different theoretical studies [6,7] on the variation of total double ionization rate with the laser intensity, were made to understand the physical mechanism involved in direct double ionization of atoms, and it has been shown [7] that the electron correlation plays an important role in direct double ionization of He. Recent experimental observations [8] on momentum distribution of recoil ions (doubly charged) were explained by considering electron correlation and field dressing of two outgoing electrons [6].

In this work, we will describe a different scenario by considering a new, non-local quantum electrodynamics, proposed by us earlier [9]. This non-local quantum electrodynamics was derived in field theoretic approach and had been shown to interpret certain features of multiphoton

\*This paper is dedicated to the memory of Professor T. K. Rai Dastidar

double ionization and above threshold ionization in an atom placed in a strong laser field. The gauge covariance of non-local electromagnetic field asserts that this non-local nature of the field is allowed by nature. It was also shown that the non-local field makes contact with the squeezed light, thus describing two completely different scenarios from a unified point of view. The non-local nature of quantum electrodynamics allows for coherent emission/absorption of two phase correlated photons by an atomic system under suitable conditions which can be met in the laboratory as well as in cosmological environments [9,10]. The most striking feature is that the phase correlated two-photon absorption/emission processes (which occurs due to the non-local nature of the field) are linear in laser intensity. It is now an experimentally observed fact that the two-photon absorption/emission processes in atoms in presence of squeezed fields are linearly dependent on laser intensity [11]. Furthermore, an arrow of time in the quantum level is obtained in order to satisfy the energy conservation in phase correlated two-photon emission/absorption processes [9]. Hence, the causality comes out as a consequence of energy conservation. Therefore, the non-local QED used in this work, has been derived from the first principle without any approximation and it has its application in diverse fields of physics [9,10,12]. Moreover, we will show below that by considering the effect of electron correlation in the matrix element of phase correlated two-photon absorption, the observed intensity dependence of DDI can be explained within the framework of perturbation theory. Furthermore, the intense field effect on outgoing electrons in the double ionization continuum can also be included by properly choosing the wavefunction of outgoing electrons.

In general, the radiation field amplitude can be described as a linear combination of its local and non-local components and we will show that the non-local component of the radiation field gives rise to direct double ionization, whereas the local component gives the sequential double ionization. In the non-local picture of the radiation field, phase correlated excitation of two electrons leads to two-photon transition, the probability of which depends linearly on laser intensity, in contrary to the local picture, where the two-photon transition probability is proportional to the square of the laser intensity. In the ladder of multiphoton transitions in an atom, if the number of photons absorbed is not sufficient to excite it to the double ionization continuum, it will single ionize and both types of transitions (due to the local and non-local component of the radiation field) will contribute depending on the laser intensity and on the phase correlated nature of the radiation field. But if the number of photons

absorbed is sufficient to double ionize the atom as mentioned before, direct and sequential double ionization will take place depending on the degree of phase correlation in the laser field and on the degree of correlation in the atomic and molecular system.

## 2. Theory

In our previous paper [9], we have derived in field-theoretic approach, the non-local radiation field and the corresponding transition amplitude due to the interaction of this non-local radiation field with the electrons. In this non-local picture, two events of photon absorption can be phase correlated, if

$$\omega\delta t \ll 1, \quad (1)$$

$\omega$  being the photon frequency and  $\delta t$  is the time gap between the two photon-absorption events. This condition can be fulfilled if the photon flux is so high that the two photons are absorbed almost simultaneously. Therefore, the necessary and sufficient condition for the phase matching of the two events is given by  $\delta t \ll \frac{1}{\omega}$  and when this condition is satisfied, two photons are coherently absorbed by the atomic electrons. For practical purposes this requirement can be replaced by a more convenient one, namely that the number of photons flowing into and out of the volume  $V$  of the atomic shell occupied by the outer electrons, in time  $\frac{1}{\omega}$ , will be much larger than the number of those outer electrons. Thus for a  $n$ -outer electron atom the condition reads

$$\frac{VI}{c\hbar\omega^2} \gg n \quad (1)$$

In conventional picture, two-photon absorption occurs (in the non-resonant case) through a virtual intermediate state of lifetime  $\sim \frac{1}{\omega}$ , which is much larger than the  $\delta t$  in (1). To describe the phenomenon of phase correlated two photon-absorption events, we have derived a non-local vector potential  $\mathcal{A}(x, x')$  in field theoretic approach [9], so that the minimal interaction at the space-time point  $x$ , is given as  $\frac{-e}{mc} \int p(x') \mathcal{A}(x, x') dx'$ , where  $x = r, t$  and  $x' = r', t'$  and the corresponding interaction matrix element is given as

$$M_{fi}(t) = \frac{-e}{mc} \langle \psi_f(x), n_f | p \cdot \mathcal{A}(x, x') | \psi_i(x'), n_i \rangle \quad (2)$$

*i.e.*, the interaction at point  $x$  is correlated with the interactions at all other points  $x'$ . Similarly the multipolar non-local interaction can be given as

$$-e \int r \cdot \varepsilon(x, x') dx' \quad (2)$$

In order to quantize this non-local field, we proceed by carrying out a Fourier expansion [13] of this non-local potential over the usual photon modes :

$$A(x, x') = \sum_{k_1 \lambda_1} \sum_{k_2 \lambda_2} [C_{12} \hat{\epsilon}_{12}(\hat{r}, \hat{r}') \exp(i\mathbf{k}_1 \cdot \mathbf{r} + i\mathbf{k}_2 \cdot \mathbf{r}' - i\omega_{k_1} t - i\omega_{k_2} t' + c.c)], \quad (3)$$

where we have written  $C_{12} = C_{k_1 \lambda_1 k_2 \lambda_2}$  and  $\hat{\epsilon}_{12} \equiv \hat{\epsilon}_{k_1 \lambda_1 k_2 \lambda_2}$  and the polarization vector  $\hat{\epsilon}(\hat{r}, \hat{r}')$  can be expressed as a power series in  $\hat{r}$  and  $\hat{r}'$  with undetermined coefficients as  $\hat{\epsilon}(\hat{r}, \hat{r}') = (\hat{r} + \hat{r}') \sum_{n=0}^{\infty} a_n P_n(\hat{r} \cdot \hat{r}')$ , which is the most general form possible. As in the standard QED, one can derive expressions for non-local field amplitude and its square from eq. (3) and hence the total field energy for the radiation field can be written as

$$W = a_I^2 W_I + a_{II}^2 W_{II}, \quad (4)$$

where  $W_I$  is the usual field energy summed over single modes and  $W_{II}$  is the non-local field energy expressed (after quantization) as :

$$W_{II} = 1/2 \sum_{k\lambda} (Y_{k\lambda}^2 + 4\omega_k^2 Z_{k\lambda}^2), \quad (4')$$

where  $k\lambda$  stands for a correlated mode pair  $k_1 \lambda_1$  and  $k_2 \lambda_2$  and  $2\omega_k$  stands for  $\omega_{k_1} + \omega_{k_2}$ .  $Y$  and  $Z$  can be expressed [9] in terms of  $C_{k\lambda}$  and  $C_{k\lambda}^*$  and satisfy the canonical equations of motion with  $W_{II}$  as the Hamiltonian as in standard QED.  $a_{II}$  and  $a_I$  are the population fractions for the correlated mode pairs and the independent modes respectively. It is to be mentioned here that to quantize the field using Coulomb gauge, we impose for the present the condition that  $(\hat{k}_1 + \hat{k}_2) \cdot \hat{\epsilon} = 0$ , but this restriction has been removed latter. By quantizing this non-local field, one can write down the field energy in terms of a new pair of creation and annihilation operators as :

$$W_{II} = 1/2 \sum_{k\lambda} \hbar \omega_k (b_{k\lambda} b_{k\lambda}^\dagger + b_{k\lambda}^\dagger b_{k\lambda}), \quad (5)$$

$$\text{where } b_{k\lambda}, b_{k\lambda}^\dagger = (2\omega_k Z_{k\lambda} \pm iY_{k\lambda}). \quad (6)$$

The two new operators obey a different commutation relation from that in standard QED :

$$\begin{aligned} [b_{k\lambda}, b_{k'\lambda'}^\dagger] &= 2\delta_{kk'} \delta_{\lambda\lambda'}, \\ [b_{k\lambda}, b_{k'\lambda'}] &= [b_{k\lambda}^\dagger, b_{k'\lambda'}^\dagger], \end{aligned} \quad (7)$$

and operates on the number states as

$$b_{k\lambda} |n_{k\lambda}\rangle = \sqrt{n_{k\lambda}} |n_{k\lambda} - 2\rangle,$$

$$b_{k\lambda}^\dagger |n_{k\lambda}\rangle = \sqrt{n_{k\lambda} + 2} |n_{k\lambda} + 2\rangle,$$

$$b_{k\lambda}^\dagger b_{k\lambda} |n_{k\lambda}\rangle = n_{k\lambda} |n_{k\lambda}\rangle.$$

Therefore,  $b$  and  $b^\dagger$  are two-photon annihilation/creation operators, and  $b^\dagger b$  is the usual number operator. Once again, by creation of two photons we mean the creation of one photon in each member of a correlated mode pair, and similarly for annihilation. One can always express the vector potential and field strengths in terms of these new operators. It is to be mentioned that the commutation relation (7) for these two new operators are gauge-independent, and hence one can do away with transversality restriction  $(\hat{k}_1 + \hat{k}_2) \cdot \hat{\epsilon} = 0$ .

Depending on the photon flux and on the coherence properties of the laser field, one must in general describe the field amplitude as

$$\epsilon = a_I E_I + a_N E_N, \quad (8)$$

where the subscripts  $L$  and  $N$  refer to local (usual) and the non-local fields respectively ( $a_L$  and  $a_N$  are identical with  $a_I$  and  $a_{II}$  in eq. (4)); relative strength of the two coefficients would depend essentially, upon the degree to which the response of the atomic electrons to the field appears phase-correlated. The first term in eq. (8) gives rise to conventional single photon transition amplitude (referred to as  $D$  in the text) and the second term gives rise to correlated two-photon transition amplitude (referred to as  $T$  in the text) (for details see eq. (14) and (19) in Ref. [9]). Formally, for two-electron atom the transition matrix element corresponding to the non-local radiation field can be written as

$$\begin{aligned} T &= \langle \psi | f, l f'(x, x'), n_{k\lambda} 2 | (r + r') \\ &\times \epsilon_N(x, x') | \psi | l, l'(x, x'), n_{k\lambda} \rangle. \end{aligned} \quad (9)$$

The time integration in (9) yields the energy balance  $\delta(E_f, E_i + 2\hbar\omega_k)$ , leaving

$$T = i \frac{\sqrt{2\pi I}}{c} \langle \psi_f(r, r') | (r + r') \cdot \hat{\epsilon}(\hat{r}, \hat{r}') | \psi_i(r, r') \rangle, \quad (10)$$

where  $E_f$  and  $E_i$  are the final and initial energy of the total system respectively and  $I$  is the photon flux. To include electron correlation, one can use the correlated wavefunctions for  $\psi_f(r, r')$  and  $\psi_i(r, r')$  (for details see Refs. [9] and [13]). To include the strong field effect on the outgoing electrons in double ionization continuum, one will have to modify the continuum correspondingly.

We will now derive the rate for double ionization in atoms and molecules considering both type of transitions  $T$  and  $D$ , within lowest order perturbation theory. In general,

this phase correlated two-photon excitation ( $T$ ) will run parallel to the conventional single photon transitions ( $D$ ) in an atom irradiated with strong radiation field and hence in any multiphoton excitation process in a field having strong second order coherence the prominence of  $T$ -type transitions or  $D$ -type transitions will depend mainly on the laser intensity. Since both the transition amplitudes  $T$  and  $D$  are proportional to  $I^{1/2}$  ( $I$  being the laser intensity), it is expected that  $T$ -type transitions in general, will be prominent in the intensity range lower than that for the  $D$ -type transitions. However there are other damping factors which will affect these two transition rates differently and hence their significance in a given intensity range may differ.

One can model a  $n$ -photon absorption process as a ladder of (i) purely  $T$ -type transitions ( $TTT\cdots$ ), (ii) purely  $D$ -type transitions ( $DDDD\cdots$ ) and (iii) a mixture of both  $T$  and  $D$ -type transitions ( $TTDDTD\cdots$  and its different variations). In a series of  $T$ -type transitions, phase correlation is maintained at each step and leads to double ejection of electrons, *i.e.* direct double ionization or non-sequential ionization occurs as long as electron correlation is maintained. The sequential ionization occurs due to purely  $D$ -type or the mixture of  $T$  and  $D$ -type transitions. In practice, all these channels will contribute and the prominence of any channel will depend on the laser intensity and other intensity dependent factors, which will be described below. Actually, the total rate will be the sum of the rates for different parallel channels. The total rate for  $n$ -photon transitions, within the lowest order perturbation theory, can be written as :

$$P_n = \sum P_i, \quad (11)$$

where  $i = 0, 2, 4, \dots, n$  for even  $n$  and  $i = 0, 2, 4, \dots, (n-1)$  for odd  $n$ .

$$P_i = P_T P_D \quad (12)$$

$$\text{where } P_T = \frac{T^{i/2}}{E_i^{i/2-1}} \quad (13)$$

$$\text{and } P_D = \frac{D^n}{E_i^{n-i-1}} \quad (14)$$

are the  $i$ -photon  $T$ -transition amplitude (for  $i \neq 0$ ) and  $(n-i)$ -photon  $D$ -transition amplitude (for  $i < n-1$ ) respectively and  $E_a$ 's are the average values of the energy denominators for non-resonant transitions.

To study the intensity variation of double ionization yields, one will have to consider the following effects on the ionization rates; (i) the effect of saturation of these rates

with the increase in laser intensity, (ii) the effect of damping due to breakdown of phase correlation, and (iii) the effect of damping due to breakdown of electron correlation. To include the effect of saturation, the rate  $P_i$ 's will be multiplied by the factor  $(1 - e^{-P_i})$ . To include the effect of damping due to breakdown of phase correlations, we have multiplied  $|P_T|^2$  by the factor  $e^{-y}$  and  $|P_D|^2$  by the factor  $(1 - e^{-y})$ , where  $y$  is given by  $2/(IV/ch\omega^2)$ . To consider the effect of breakdown of electron correlation we have multiplied  $|P_T|^2$  by  $e^{-\sqrt{II_c}}$ , where  $I$  is the laser intensity and  $I_c$  is the laser intensity at which the electric field due to the laser radiation is equal to the atomic potential. Hence, the total rate in this model can be given as :

$$P_i = \sum P_i (1 - e^{-P_i}) (1 - e^{-y}) e^{-y} e^{-\sqrt{II_c}} \quad (15)$$

where  $i = 0, 2, 4, \dots, n$ , for even  $n$  and  $i = 0, 2, 4, \dots, (n-1)$ , for odd  $n$ . It is to be noted that purely  $D$ -type transitions *i.e.*, for  $i = 0$ , the factor  $e^{(-y-\sqrt{II_c})}$  will be omitted and for purely  $T$ -type transitions *i.e.*, for  $i = n$ , the factor  $(1 - e^{-y})$  will be omitted. To obtain rates for  $n$ -photon ionization using this model, the parameters  $T$ ,  $D$  and  $i$  (which have been defined above from the first principle), have to be chosen by considering the physical conditions. Details of these parameters have been discussed in the next section. Moreover, the intensity  $I_c$  has been defined as the intensity for which the magnitude of the radiation field is equal to that of the coulomb field within the atom. It is well known that the electron correlation in an atom depends on the Coulomb field between the electrons. In case of interaction of radiation field with the outer shell electrons of the atom, electron correlation between outer shell electrons will affect the photon absorption process. Therefore, if the radiation field is strong enough to dominate over Coulomb field between the outer shell electrons, electron correlation will breakdown. The magnitudes of these Coulomb fields depend on the radius of the outer shell electrons and for atoms of high nuclear charge, outer electrons will occupy the shell of high principal quantum number. Hence, the threshold intensity at which these Coulomb fields will breakdown, depends on the nuclear charge of the atom being ionized and the principal quantum number of the shell from which it is being ionized. Choosing a field of order  $10^{-2}$  a.u., a crude estimate for an upper limit  $J_{\max} \sim 10^{13}$  W/cm<sup>2</sup> to the laser intensity can be made up to which we can expect electron correlation in a two-electron atom to be maintained.

### 3. Results and discussion

In this work, the results for  $n = 6$  have been shown. For  $n = 6$ , four channels will contribute, where the channel

amplitudes will be proportional to (i) *TTT*, (ii) *TTDD*, (iii) *TDDDD* and (iv) *DDDDDD*. The channel (i) which corresponds to purely *T*-type transitions, gives rise to non-sequential ionization, the channel (iv) which corresponds to purely *D*-type transitions will give rise to the sequential ionization. The other two channels which are the mixture of *T* and *D*-type transitions will give rise to sequential or non-sequential ionization, depending on the number of *D*-transitions present in the ladder of excitation. The presence of too many *D*-transitions will break down the phase correlation, leading to sequential ionization. In this ladder of transitions, the values of *T* and *D* may vary in each consecutive steps, but we have replaced the *T*'s and *D*'s by their average values. Moreover, the energy denominators involved in each non-resonant step of transitions have been replaced by an average value. We have shown here results of parametric calculations for 6-photon non-resonant transitions.

In Figure 1, the laser intensity variation of four individual channels, together with the total has been shown. It is

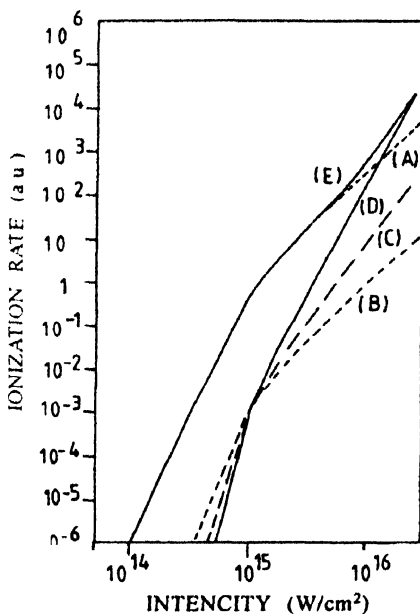


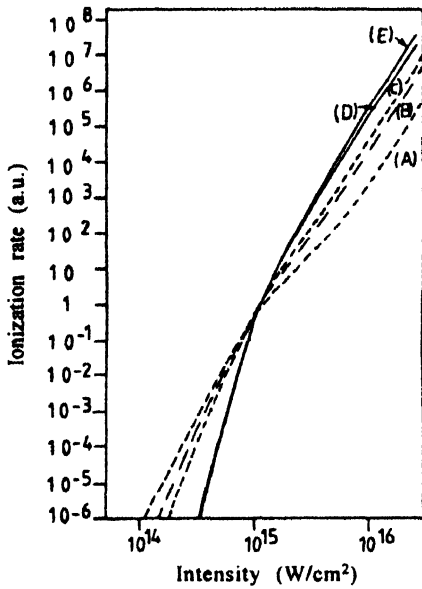
Figure 1. Variation of double ionization rates with laser intensity. Curves (A) rate for channel (i) : *TTT*; (B) rate for channel (ii) : *TTDD*, (C) rate for channel (iii) : *TDDDD*; rate for channel (iv) : *DDDDDD* and (E) Total : sum of these four rates.  $\lambda = 780$  nm, number of photons  $n = 6$ . Volume of the atomic shell  $V = 10^6$  a.u.,  $|T_\mu|^2 = 30$  a.u. and  $|D_\mu|^2 = \sqrt{800}$  a.u. (see text).

found that with the increase in laser intensity, initially the NS channel dominates over all the other channels and the kink in the total rate appears at the laser intensity, where the saturation of the NS channel starts. Towards higher intensity, the total rate is dominated by the sequential channel and finally it follows the intensity variation of sequential channel.

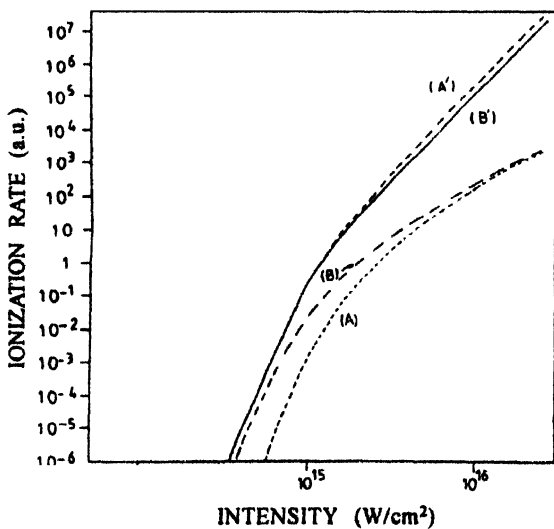
At the intermediate intensities, it is found that the contribution from the NS channel is orders of magnitude greater than that from the sequential channel. All these features have been observed [5] in the intensity variation of double ionization of He atom. The intensity variation of double ionization rate shown in Figure 1, resembles that of the observed double ionization yield for the He atom (see Figure 1 of Ref. [5]). In this calculation, we have chosen  $|T|^2 = |T_\mu|^2 I$  where  $|T_\mu|^2 = 30$  a.u. and  $|D|^2 = |D_\mu|^2 I$  where  $|D_\mu|^2 = \sqrt{800}$  a.u. In case of dipole transitions,  $T_\mu$  is the phase correlated two photon dipole transition moment due to the interaction of atom with the non-local component of the electromagnetic field and  $D_\mu$  is the conventional dipole transition moment due to the interaction of the atom with the local component of the electromagnetic field. As mentioned above, details of calculation of  $T_\mu$  and  $D_\mu$  have been discussed in Ref. [9]. For this calculation, the wavelength  $\lambda = 780$  nm and volume  $V = 10^6$  a.u. Hence, the radius of the atomic shell from which the ionization takes place is 62.035 a.u. the radius of the atomic shell can be large when the ionization takes place from an excited state. We have taken average value of the energy denominator  $E_a = 1$  a.u. throughout this calculation. For other values of  $E_a$ , transition rates  $P_T$ ,  $P_D$  and  $P_i$ 's given here, will be scaled accordingly.

As mentioned before, the value of  $y$  is given as the ratio of number of outer shell electrons of the atom and the number of photons flowing into and out of the volume  $V$  of the atomic shell occupied by the outer electrons in time  $1/\omega$  and hence the damping of the ionization process will depend on the availability of photons compared to the number of electrons on the outer shell. Therefore, the value of  $y$  (see also the expression for  $y$  in 'Theory') depends on the (i) volume  $V$  of the atomic shell from which the ionization occurs, (ii) the frequency of the laser radiation and (iii) the intensity of the laser *i.e.* the photon flux. Hence, the damping of the ionization yield will also depend on these three factors. To show the influence of the wavelength of the laser radiation and the volume  $V$  of the atomic shell (from which the ionizing transition occurs) on the double ionization, we have repeated the calculation with  $\lambda = 248$  nm and for different values of  $V$ , with the values of  $T$  and  $D$  being the same as in Figure 1. For this wavelength, since the value of  $\omega$  increases, the value of  $y$  also increases, giving rise to damping of the NS channel. But this damping effect is more prominent when the value of  $V$  is decreased. For this wavelength ( $\lambda = 248$  nm), since the photon energy is more than three times larger than that for radiation with wavelength 780 nm, and the number of photons absorbed for the

ionizing transition is the same ( $n = 6$ ), the 6-photon ionizing transition will occur from a low energy state of the atom. *i.e.* the volume  $V$  will be less than that used for the calculation with  $\lambda = 780$  nm. Hence, for this wavelength, we have plotted the total double ionization rate as a function of laser intensity, for different values of  $V$  (Figure 2). We find that as the volume  $V$  decreases, the double ionization rate starts contributing at higher and higher laser intensities. This is because of the fact that as the volume decreases the value of  $y$  increases and the damping on the NS channel (*i.e.*



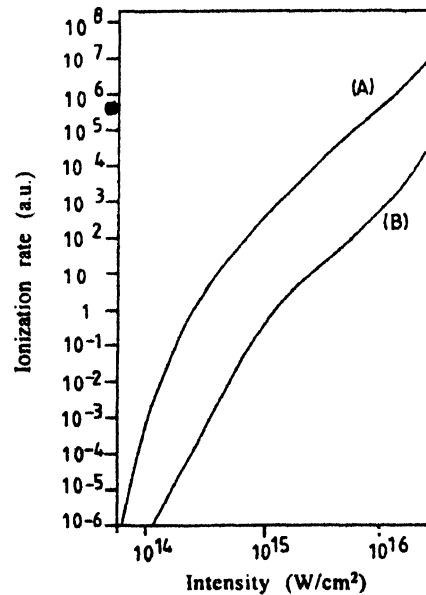
**Figure 2.** Total double ionization rate as a function of laser intensity for different values of  $V$ . Curves : (A) for  $V = 10^6$  a.u.; (B) for  $V = 10^5$  a.u.; (C) for  $V = 5.10^4$  a.u.; (D) for  $V = 10^4$  a.u.; (E) for  $V = 5.10^3$  a.u.  $\lambda = 248$  nm, other parameters are the same as in Figure 1.



**Figure 3.** Intensity variation of rates for two channel (i) TTT and (iv) DDDDDD for two values of  $V$ . Curves : (A) rate for NS channel (TTT) and (B) rate for sequential channel (DDDDDD), for  $V = 5.10^3$  a.u.; (A') rate for NS channel (TTT) and (B') rate for sequential channel, for  $V = 10^4$  a.u. Other parameters are the same as in Figure 2.

purely  $T$ -transitions) increases. As a result, NS double ionization rate becomes appreciable at the higher values of laser intensity for the lower values of  $V$ , and the total rate shows no kink like structure in the intensity variation. To demonstrate this, we have plotted the NS rate (purely  $T$ -transitions) and the sequential rate (purely  $D$ -transitions) for two values of  $V$ ,  $V = 10^4$  a.u. and  $V = 5.10^3$  a.u., in Figure 3. Values of  $T$  and  $D$  are the same as in Figure 2. It is found that the NS rate is strongly damped and becomes significant only at high laser intensity and almost coincides with the sequential rate for  $V = 10^4$  a.u.; whereas for  $V = 5.10^3$  a.u., NS rate is appreciable at much higher intensity than that for the sequential rate (purely  $D$ -transitions). Hence, in this case, total double ionization rate is dominated by the sequential ionization, the non-sequential ionization being negligible in this intensity range. Therefore, for shorter wavelength, non-sequential ionization becomes weak and the sequential ionization dominates, which is also an observed feature [4], in double ionization of atoms.

If we consider the single ionization of the atom by the absorption of same number of photons (*i.e.*  $n = 6$ ), and for the wavelength  $\lambda = 780$  nm, the 6-photon ionizing transition will occur from much lower energy state than that for double ionization. Hence, the value of  $V$  will be much less than that used for the calculation of double ionization rate. Moreover, the single ionization rate is orders of magnitude greater than the double ionization rate. Hence, by increasing the value of  $|T_{\mu}|^2 (= 300$  a.u.) and decreasing the value of  $V (= 5.10^3$  a.u.), we obtained a single ionization rate, where the kink in the curve is almost absent (Figure 4). This feature has



**Figure 4.** Intensity variation of single and double ionization rates. Curve (A) : Single ionization rate,  $V = 5.10^3$  a.u.,  $|T_{\mu}|^2 = 300$  a.u.. Curve (B) : Double ionization rate,  $V = 10^6$  a.u.,  $|T_{\mu}|^2 = 30$  a.u. Other parameters are the same as in Figure 1.

been observed for the single ionization of He atom (Figure 1 of Ref. [5]). To compare the single ionization rate with that for double ionization, we have also plotted here the double ionization rate (shown in Figure 1) and it is found that the curves in Figure 4 resembles those of Figure 1 of Ref. [5].

In conclusion, we have shown that the non-local nature of the radiation field can give rise to phase correlated excitation of two electrons, leading to direct double ionization (or non-sequential double ionization) and this phase correlated two-photon rate depends linearly on laser intensity. Combining this rate with the sequential ionization rate (which comes in due to the local nature of the radiation field) can reproduce similar intensity dependence of double and single ionization yields, as observed in the experiments [4,5] on single and double ionization of He atom. Furthermore, we have demonstrated the physical basis for wavelength dependence of non-sequential double ionization of atoms.

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