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Coulomb effect in multiphoton ionization of rare-gas atoms

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Abstract. Using 200 fs, 800 nm Ti:sapphire laser pulses to ionize noble atom gases, the ion versus intensity curves of these gases were obtained. By comparing the experimental curves with those obtained using different theoretical ionization rates, it was found that the inclusion of the effects on the ionization rate of the long-range Coulomb potential of the parent ion through quasi-classical perturbation theory (as proposed by Perelomov *et al* and Krainov) gives a rather good overlap with the experimental results.

1. Introduction

Recent extensive studies of the phenomenon of the interaction of ultra-fast strong laser fields with atoms and molecules has shown that multiphoton ionization (MPI) to singly charged ions, apart from its intrinsic interest, is essential in the discussion of phenomena such as non-sequential ionization (Walker *et al* 1994, Kuchiev 1995, Talebpour *et al* 1997a), high-order harmonic generation (Kulander and Shore 1989, Levenstein *et al* 1994, Chin and Glovinski 1994) and fragmentation of diatomic molecules (Talebpour *et al* 1997b). This importance requires a satisfactory theory for predicting the MPI of atoms and molecules. In the quasi-static regime, the task is adequately fulfilled by the ADK theory (Ammosov *et al* 1986). Presently, only experiments using CO_2 lasers (Walsh *et al* 1994, Chin *et al* 1985) strictly occur in this regime, although experiments using a 1.053 μm laser (Augst *et al* 1991, Auguste *et al* 1992) could also be adequately described by the ADK theory. However, most other experiments using visible or near-infrared lasers take place in the multiphoton or intermediate regime, where the ADK model underestimates the ionization rate. Another model is thus necessary for describing the MPI in the intermediate regime.

Since 1965, a vast amount of theoretical analysis within the framework of the KFR theory (Keldysh 1965, Faisal 1973, Reiss 1980) has been carried out on the formulation of a predictive analytical model for multiphoton ionization in strong laser fields. Two of these, interesting in part due to their simplicity, are the strong-field approximation (SFA, Reiss 1980) and Szoke's model (Perry *et al* 1988). These two models, although successful in providing many useful qualitative predictions, have not been able to provide a quantitative fit with the experimental data (see below) due to their shortcoming in handling the Coulomb potential of the parent ion: the SFA model completely neglects the Coulomb effect in the final state, while Szoke's model considers the Coulomb potential, in the final state, as a constant. Recently, Krainov (1997) has succeeded in a rather simple way to include the effect of the Coulomb potential of the parent ion on the ionization rate. He showed that,

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with the correction thus made, the rate predicted by this model asymptotically approaches the rate of the ADK theory in the quasistatic limit $\gamma \ll 1$. ($\gamma = \sqrt{E_i/2U_p}$ is the Keldysh parameter (Keldysh 1965). In this formula, E_i is the ionization potential and U_p is the ponderomotive energy.) This is an essential criterion for hoping to have a predictive theory and this is not satisfied by the SFA and Szoke's model. Krainov's approach is rather similar to that proposed by Perelomov and Popov (1967) and Perelomov *et al* (1966) and the final rates of these two theories agree as will be shown (Perelomov, Popov and Terent'ev's model (1966) will be referred to as the PPT model). Other approaches for including the Coulomb field have also been proposed which successfully explain some aspects of the ATI ionization (Kamiński and Ehlotzky 1997, Faisal and Becker 1997). They, however, require much more complex calculations which are normally beyond the capacity of experimental laboratories.

Our goal in the present paper is to show that PPT and Krainov's models rather accurately predict the total ionization rate of atoms in the intermediate regime where the ionization parameter γ is of the order of 1. We will compare the ion versus intensity curves predicted by five different models (SFA, Szoke, Krainov, PPT and ADK) with the experimentally obtained ion yield of rare-gas atoms measured in a femtosecond Ti:sapphire laser field. This comparison is made over a dynamic range of seven to eight orders of magnitude in the ion yield.

2. Experimental results and discussion

We have studied the MPI of Xe, Kr, Ar, Ne and He using a stable, Ti:sapphire laser operating at 800 nm, with a pulse length of 200 fs. A complete description of the experimental technique can be found in Talebpour *et al* (1996). Briefly, the laser was focused using $f/100$ optics into an UHV chamber having a background pressure of 2×10^{-9} Torr. Ion species were separated with a time-of-flight mass spectrometer. Ion curves were produced by combining a series of intensity scans, each having a different fill pressure in the interaction chamber. The gas pressure ranged from 10^{-8} to 10^{-4} Torr.

The absolute intensity calibration is based on a comparison with the saturation intensity predicted by the ADK formula in the case of helium. As has already been observed (Walker *et al* 1994), near the saturation region, the experimentally measured He^+ signal in a Ti:sapphire laser field has an excellent overlap with the ion yield predicted by the ADK theory. Intensity calibration of other curves is established in comparison with that of He and is thus free of any assumption on the validity of the ADK model for these gases. The calibration technique used in these cases was to scan successively the saturation part of all gases without, between two runs, changing the experimental conditions. The saturation intensity of each gas could then be obtained in comparison with that of He, thus establishing the calibration. The accuracy in the relative intensity calibration between one given ion curve and the reference He curve is estimated to $\pm 5\%$ (see figure 1).

The experimentally measured ion yield versus intensity curves were compared with those predicted by five models: ADK, SFA, PPT, Szoke's and Krainov's models. In each case the theoretical ion curve is calculated by integrating the rate equation, taking into account the spatial and temporal dependence of the intensity, and consequently the ionization rates (Chang *et al* 1992). The absolute ion signal is not considered: due to the kind of detector used (electron multiplier tube), it is very difficult to know the precise number of ions detected in each shot. Consequently, it seems preferable to match the saturation signals of experimental and theoretical curves because this does not depend on any laser parameter. The effect of the multiply charged ions is considered negligible in this comparison. As some previously published data show (Talebpour 1997a), before saturation

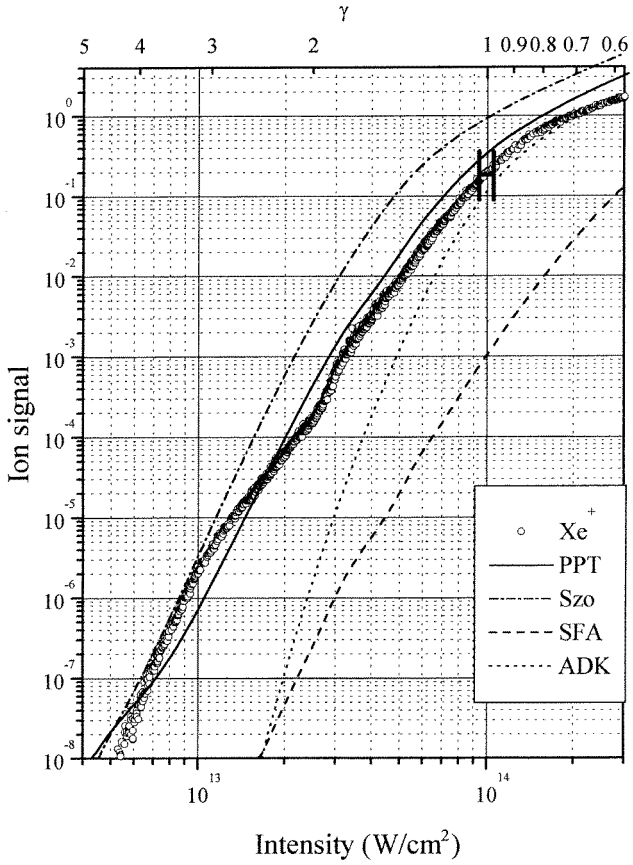

Figure 1. Multiphoton ionization of Xe using stable linearly polarized laser pulses from a Ti:sapphire laser (800 nm). Each datum corresponds to a three-point average. The upper scale is the γ scale (Keldysh parameter) for Xe. The error bar (|–|) near the saturation part of the experimental Xe^+ curve is the experimental error in the relative intensity calibration of Xe compared to the He curve. The theoretical ion yields are, from left to right, calculated from Szoke's model, PPT model, ADK theory and SFA. The theoretical curve calculated using Krainov's model grossly overlaps with the PPT curve and is thus not shown.

of the first charge, these multiply charged ions do not contribute more than 2% or 3% to the total number of ions.

For an atom with ionization potential E_i, effective principal quantum number n^*, orbital angular momentum l and magnetic quantum number m in a laser field of frequency ω, the ADK model gives the following total ionization rate:

$$W_{m\mathrm{ADK}} = |C_{n^*l^*}|^2 f_{lm} E_i \sqrt{\frac{6}{\pi}} \left(\frac{2(2E_i)^{3/2}}{F} \right)^{2n^*-|m|-3/2} \exp\left(-\frac{2(2E_i)^{3/2}}{3F} \right) \tag{1}$$

where F is the electric field of the laser and the factors f_{lm} and $C_{n^*l^*}$ are, respectively,

$$f_{lm} = \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!} \tag{2}$$

$$|C_{n^*l^*}|^2 = \frac{2^{2n^*}}{n^*\,\Gamma(n^*+l^*+1)\,\Gamma(n^*-l^*)}. \tag{3}$$

For ionization rate calculations, we use $l^* = n^* - 1$, the ground state values for n^* and l. We average over the possible values of m:

$$W = \frac{1}{2l+1} \sum_{m=-1}^{l} W_m. \tag{4}$$

The total rate of ionization in SFA and Szoke's model are given by (5) and (6), respectively,

$$W_{\mathrm{SFA}} = \sum_{n=N}^{\infty} 2\pi\omega^2 p(n - n_{\mathrm{osc}})^2 \int \mathrm{d}\Omega \, |\Phi(\boldsymbol{p})|^2 \, J_n^2(n_{\mathrm{f}}, -n_{\mathrm{osc}}/2) \tag{5}$$

$$W_{\mathrm{SZO}} = \sum_{n=N}^{\infty} 2\pi\omega^2 p \frac{n^2(n - n_{\mathrm{osc}} - n_{\mathrm{b}})}{n - n_{\mathrm{osc}}} \int \mathrm{d}\Omega \, |\Phi(\boldsymbol{p})|^2 J_n^2(n_{\mathrm{f}}, -n_{\mathrm{osc}}/2) \tag{6}$$

where N is the minimum number of photons necessary to ionize the atom, $n_{\mathrm{b}} = E_{\mathrm{i}}/\omega$, $n_{\mathrm{osc}} = U_{\mathrm{p}}/\omega$ (U_{p} is the ponderomotive energy), $n_{\mathrm{f}} = 2[n_{\mathrm{osc}}/\omega]^{1/2} p\cos(\theta)$, $\Phi(\boldsymbol{p})$ is the normalized momentum space wavefunction of the electron in the ground state and $J_n(u, v)$ is the double Bessel function (Reiss 1980). In equation (5), $p = [2\omega(n - n_{\mathrm{osc}} - n_{\mathrm{b}})]^{1/2}$ while, in equation (6), $p = [2\omega(n - n_{\mathrm{osc}})]^{1/2}$. θ is the angle between $\boldsymbol{p}$ and the polarization of the laser.

As in Szoke's model, Krainov's model is also based on SFA theory. However, instead of introducing a constant potential to take into account the Coulomb field, it considers the correction to the final state wavefunction through a propagator of the electron in the Coulomb field which is obtained within the framework of the quasi-classical approximation. The resulting rate for linear polarization is

$$W_{\mathrm{KRA}} = \sum_{n=N}^{\infty} 2\pi\omega^2 p(n - n_{\mathrm{osc}})^2 \int \mathrm{d}\Omega \, |\mathrm{FT}(I_{\mathrm{KRA}}\Phi(\boldsymbol{r}))|^2 \, J_n^2(n_{\mathrm{f}}, -n_{\mathrm{osc}}/2) \tag{7}$$

where FT is the three-dimensional Fourier transform and I_{KRA} is the Coulomb correction introduced in the final state. Other symbols have the same significance as in the SFA model. The value of I_{KRA} is

$$I_{\mathrm{KRA}} = \left(\frac{2Z^2}{n^{*2}Fr}\right)^{n^*}. \tag{8}$$

In this formula, F is the amplitude of the electric field and r is the radial coordinate. In the calculations of the rate for SFA, Szoke's and Krainov's models, we tried two different sets of wavefunctions: hydrogenic wavefunctions (Nyden Hill 1996) and asymptotic wavefunctions (Ammosov *et al* 1986). These wavefunctions are, respectively, scaled with Z^* and n^* in order to obtain the correct ionization potential ($E_{\mathrm{i}} = Z^{*2}/2n^{*2}$). It was found that the asymptotic wavefunction with quantum number $l = 0$ gives the best results. Finally, the three-dimensional Fourier transform is calculated using Hankel transform (Hoang Binh and Van Regemorter 1997).

The PPT model was originally derived for a short-range potential and includes the effect of the long-range Coulomb interaction through the first-order correction in the quasiclassical action (Perelomov and Popov 1967). This approach is similar to the one followed by Krainov; however, their correction factor is different:

$$I_{\mathrm{PPT}} = \left(\frac{2(2E_{\mathrm{i}})^{3/2}}{F}\right)^{n^*}. \tag{9}$$

The rate obtained (including the Coulomb correction I_{PPT}^2) is

$$W_{n\mathrm{PPT}} = |C_{n^*l^*}|^2 f_{lm} E_{\mathrm{i}} \sqrt{\frac{6}{\pi}} \left(\frac{2(2E_{\mathrm{i}})^{3/2}}{F} \right)^{2n^* - |m| - 3/2} (1+\gamma^2)^{|m|/2+3/4} \times A_m(\omega, \gamma) \exp\left(-\frac{2(2E_{\mathrm{i}})^{3/2}}{3F} g(\gamma) \right) \tag{10}$$

where

$$g(\gamma) = \frac{3}{2\gamma} \left[\left(1 + \frac{1}{2\gamma^2} \right) \sinh^{-1}(\gamma) - \frac{\sqrt{1+\gamma^2}}{2\gamma} \right]. \tag{11}$$

The coefficients A_m can be found in Perelomov *et al* (1966) and Ilkov *et al* (1992). Some more precise values of coefficients α, β and ρ, which enter into the calculation of A_m, can be found in Perelomov *et al* (1968). The coefficients found in the latter paper involve other Coulomb corrections; however, they only have a very weak effect on the ionization rate. The coefficients f_{lm} and $C_{n^*l^*}$ are given, respectively, by (2) and (3). As for ADK, this rate should be averaged over the different values of m.

For all the cases considered here, rates predicted by PPT and Krainov's models, the latter using an asymptotic wavefunction with $l = 0$, are nearly equal (for unpolarized atoms, the PPT model is independent of l because, in the sum over m, only the term with $m = 0$ contributes). However, two differences between these models can be pointed out: firstly, the PPT model, in terms of ease of calculation, is superior to Krainov's model mainly because it avoids the use of double Bessel functions (this is, however, at the expense of more approximations). Secondly, the derivation of Perelomov *et al* seems to be more coherent than that of Krainov. PPT only considers the residual Coulomb correction at large distances from the nucleus where the Coulomb field creates a weak perturbation on the quasi-classical path of the electron. Krainov, on the other hand, integrates the classical action from the initial place of birth of the ionized electron.

Interestingly, the derivation in the S-matrix formalism of the PPT rate (i.e. using the PPT hypothesis) gives exactly the same result as obtained by Krainov. Indeed, the asymptotic initial wavefunction used by PPT is

$$\Psi(\boldsymbol{r}) = C_{n^*l^*} \left(\sqrt{2E_{\mathrm{i}}} \right)^{3/2} \left(r\sqrt{2E_{\mathrm{i}}} \right)^{\lambda - 1} \mathrm{e}^{-r\sqrt{2E_{\mathrm{i}}}} Y_{lm}(\boldsymbol{r}/r) \tag{12}$$

where $\lambda = 0$ for the short-range potential ($\lambda = n^*$ is the Coulomb asymptotic wavefunction used by Krainov). For the derivation, the final state is chosen as a Volkov state which is an asymptotically valid limit for this short-range potential. Considering the Coulomb correction of PPT (equation (9)), the final rate will be similar to that of Krainov's model except for the argument of the Fourier transform, $I\Psi(\boldsymbol{r})$. But, because $\Psi_{\lambda=0} I_{\mathrm{PPT}} = \Psi_{\lambda=n^*} I_{\mathrm{Kr}}$, the two formulae become identical. Note that the equivalence of this last derivation with PPT's restores the gauge independence (calculations of PPT are performed in the E-gauge, while this last derivation is in the A-gauge).

In figures 1–5 we present the ion yields versus peak intensity (lower horizontal scale) or the corresponding γ scale (upper horizontal scale) of xenon, krypton, argon, neon and helium, respectively. On each graph, the experimentally measured curve is compared with four of the five previously discussed theoretical rates (as mentioned previously, Krainov's and the PPT model are nearly equivalent. Only the latter is presented on the graphs). Firstly, it can be noted that the slopes predicted by the ADK theory overestimate those observed experimentally, especially for the low ionization potential gases (Xe and Kr). Also, agreement between experiment and ADK is observed only in the upper part of the ion yields of He and Ne. This is expected since the γ parameter is less than 0.5 in this

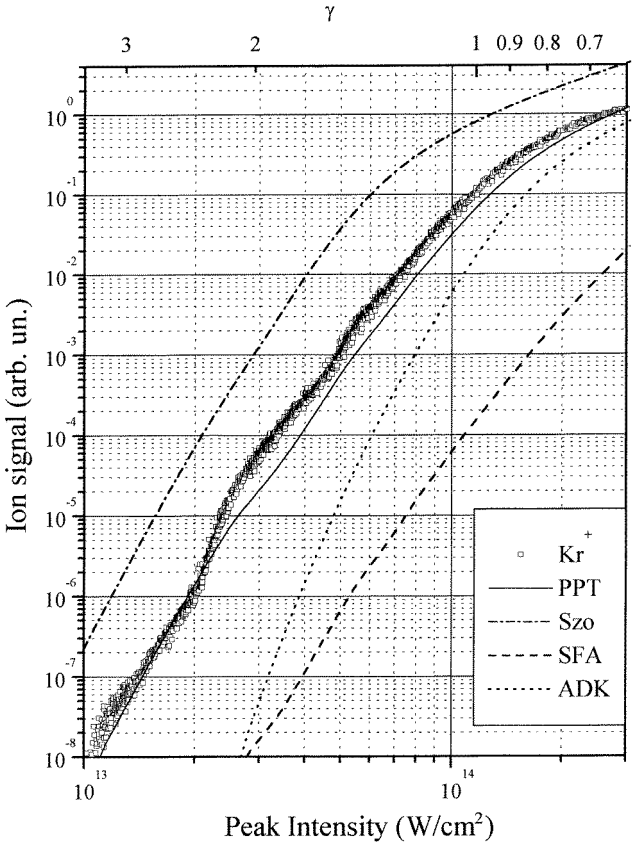

Figure 2. Same as figure 1, for Kr.

region. As concluded by Ilkov *et al* (1992), use of the ADK theory should be limited to regions of intensity where γ is smaller than 0.5.

On the other hand, the three other theories predict, in each case, the experimentally observed slope, i.e. the linear part (on a log–log plot) of each theoretical curve is almost parallel to the corresponding part of the experimental curve (this property of predicting the correct slope for a given atom is not necessarily observed in all Keldysh-type models. For example, as can be seen from figure 1 of Kamiński *et al* (1996), their model predicts an increase of the ionization rate for the H atom which is far inferior to the one predicted by the SFA (Gordon–Volkov final state) which, in view of the present analysis, is expected to predict this increase correctly). However, the SFA theory underestimates strongly (between two and three orders of magnitude) the ionization rate for all gases. The problem encountered by the SFA theory could not be solved by using a hydrogenic-like initial wavefunction instead. (Different wavefunctions of hydrogen with Z adjusted to scale the ionization potential of the atom were tried. None of them increase the theoretical rate strongly.) Finally, Szoke's model only has poor agreement with the experimental curve for Xe. For other gases, the ionization rate is strongly overestimated. Actually, the rate predicted by Szoke's model seems to be rather insensitive to the ionization potential.

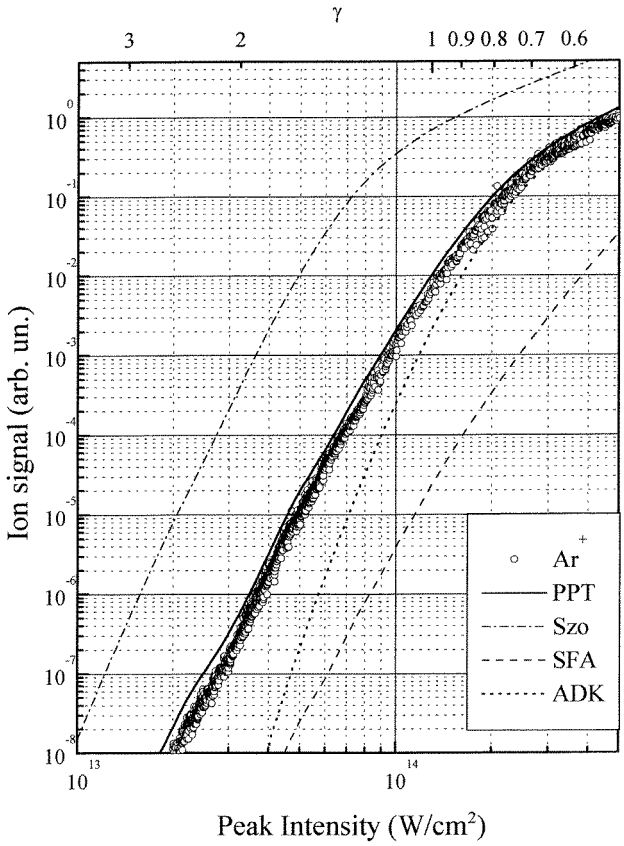

Figure 3. Same as figure 1, for Ar.

The PPT model predicts accurately the ionizing rates of He and Ne, while for Ar it is slightly overestimated. For Xe and Kr, the deviation is larger. Still, the theoretically predicted and the experimentally observed rates agree to within a factor 2. The structures that can be observed in the ion yields of Xe and Kr are not reproduced by the PPT theory. Previously, these structures were considered as an indication of a possible dynamic trapping (Talebpour *et al* 1996) and some recent numerical simulations taking into account this mechanism have been able to fit our experimental ion yield of Xe (Kulander 1997). Obviously, the PPT model, which does not consider any discrete level other than the ground state, cannot be expected to predict these effects. The only suppression of the rate that this theory can predict is due to channel closing.

Apart from neglecting the discrete levels, the other main limitation to the applicability of the PPT model is the accuracy of the Coulomb correction. An analysis made by Perelomov *et al* (1968) shows that this correction should be accurate if $\sigma(\gamma) \ll \sigma_{\rm cr}$ where

$$\sigma(\gamma) = \gamma/\gamma_k \tag{13}$$

$$\gamma_k = \frac{2E_{\rm i}}{\lambda\omega} \tag{14}$$

and $\sigma_{\rm cr}$ is defined as the limiting value of σ for which the following equation has a real

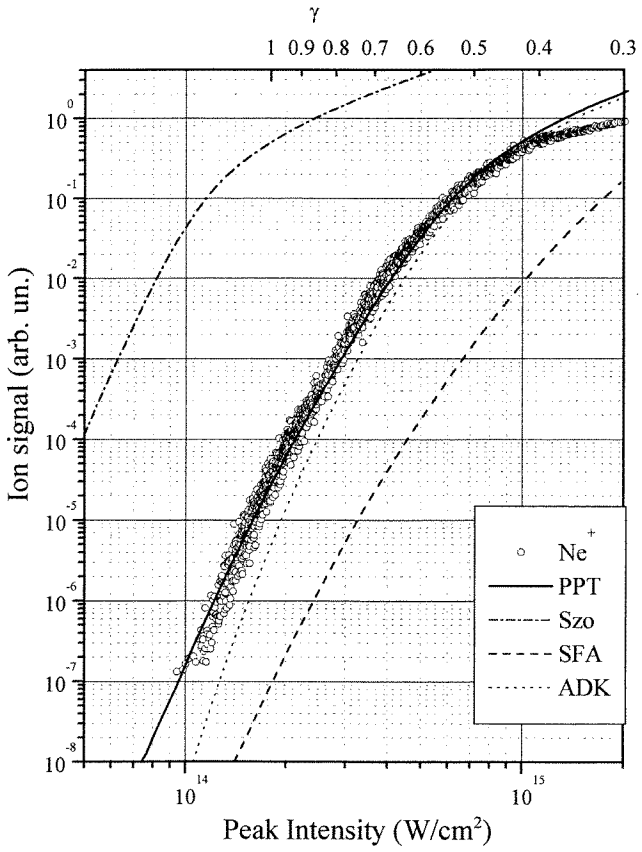

Figure 4. Same as figure 1, for Ne.

Table 1. Critical γ for different gases at which the Coulomb correction should break down.

Gases	γ_{cr}	γ_k
Xe	5.8	14.8
Kr	8.3	18.3
Ar	9.4	21.9
Ne	9.8	35.1
He	10.0	42.6

solution for τ:

$$\frac{1}{\gamma}\left(\cosh(\tau) - 1 - \tfrac{1}{2}\tau(\sinh(\tau) - \gamma)\right) = \left(\frac{\tau\sigma}{\sinh(\tau) - \gamma}\right)^{1/2} \tag{15}$$

($\lambda = n^*$ is the long-range correction to the asymptotic wavefunction). This criterion is equivalent to $\gamma \ll \gamma_{cr}$ where $\gamma_{cr} = \sigma_{cr}\gamma_k$. The values of γ_{cr} and γ_k for the different gases are presented in table 1. From this and figures 1–5, it can be seen that the experimental value of γ is much smaller than γ_{cr} for He and Ne. Consequently, it can be said that the Coulomb correction is accurate enough for the present experimental conditions. Interestingly, even with $\gamma < \gamma_{cr}$ for the other three gases, the fit is still reasonably good.

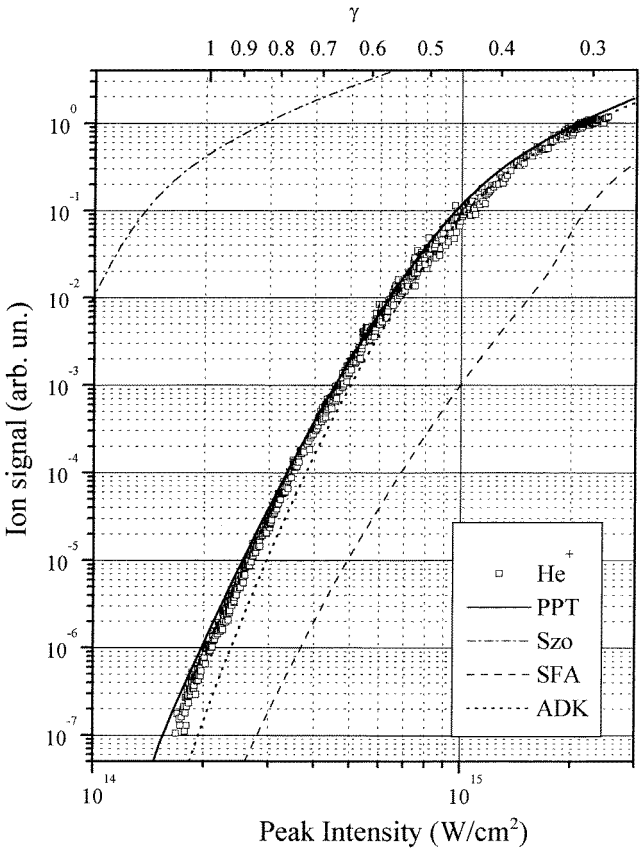

Figure 5. Same as figure 1, for He.

In conclusion, it was shown that the total ionization rate predicted by the PPT model and Krainov's model fit rather accurately the experimental ion yields for all rare gases using 800 nm femtosecond laser pulses in the intermediate regime ($\gamma \sim 1$). This contrasts with the SFA model which strongly underestimates the rate of ionization and with the ADK model, which only gives a rather good saturation intensity but not an acceptable behaviour for the ion yield. The PPT model seems to be a useful reference for comparing total rates observed theoretically and experimentally.

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