

Post ionization alignment of the fragmentation of molecules in an ultrashort intense laser field

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Abstract

We studied the angular distributions of the fragmented ions of diatomic molecules in an intense linearly polarized short laser pulse. In addition to the well-known dynamic alignment of the neutral molecules before ionization, we identified a more important post ionization alignment effect of the molecular ions. The latter is modelled quantum mechanically as resulting from the breakup of a rotating linear rotor. We showed that only for very short pulses are the two alignment mechanisms not important. In this case the angular distributions of the fragmented ions mimic the shape of the electronic density of the outermost molecular orbital.

(Some figures in this article are in colour only in the electronic version)

When neutral molecules are exposed to an intense laser pulse, they can be aligned as well as ionized by the electric field of the laser (Stapelfeldt and Seideman 2003). The coexistence of both effects makes it difficult experimentally to extract the dependence of ionization rates on the alignment of molecules with respect to the direction of laser polarization. Recently, Litvinyuk *et al* (2003) exploited the pump–probe technique on N₂ molecules where a weak laser pulse was used to align the rotational wave packet and a strong probe pulse was used to determine the ionization rates of these aligned molecules. Their results are in general agreement with the prediction of the molecular tunnelling ionization theory (Tong *et al* 2002), as well as with the interference model (Muth-Bohm *et al* 2000, Kjeldsen and Madsen 2004) in the strong field approximation, that the ionization rates peak at $\theta = 0$ (θ is the angle between the molecular axis and laser field polarization direction) for N₂ molecules. A similar measurement on O₂ was inconclusive, however. More recently Légère (2004) has shown that the harmonic generation from O₂ peaks when the molecule is not aligned along the polarization axis, again in agreement with the molecular tunnelling ionization theory.

According to the molecular tunnelling ionization theory (MO-ADK) (Tong *et al* 2002), the alignment dependence of the ionization rates of molecules by an intense laser pulse depends critically on the symmetry of the outermost orbital of the molecule. For N₂, it

is a $3\sigma_g$ orbital and the ionization rate should peak at $\theta = 0$ where the molecular orbital has the maximum density along the electric field direction of the laser. For O_2 , it is a π_g orbital and the ionization rate should peak near $\theta = 40^\circ$ (Zhao *et al* 2003, Jaron-Becker *et al* 2004). At this angle, the π_g orbital would have the largest electron density along the electric field direction of the laser. The prediction of the MO-ADK theory is very general and it is desirable that such predictions be tested for a large sample of molecules at the quantitative level. However, to deduce the alignment dependence of the ionization rates experimentally, one needs to be able to disentangle other effects that align the molecules by the laser itself (Dietrich *et al* 1993, Posthumus *et al* 1998). In this paper, we report a study on two alignment effects. One is the familiar dynamic alignment (DA) before the molecule's breakup (Stapelfeldt and Seideman 2003), the other is the new post-ionization alignment (PIA) effect which occurs after the molecules are ionized. The PIA is similar to the molecular reorientation during the dissociative multiphoton ionization which was observed by Dietrich *et al* (1993). Here we improve their model by (1) describing the rotational motion by the Schrödinger equation instead of the classical equation of motion; (2) using the 'realistic' polarizability as a function of the internuclear separation calculated by *ab initio* quantum chemistry code (GAMESS) (Schmidt *et al* 1993); (3) separating and calculating quantitatively the DA and the PIA effects. We chose O_2 as an example since according to the MO-ADK theory the laser ionization favours molecules aligning at an angle about $\theta = 40^\circ$ from the polarization axis, while both DA and PIA try to reorient the molecules and the fragment ions into the direction of the laser polarization. The shift of angular distributions of the resulting ions to the field polarization direction measures directly the alignment effect due to DA and PIA. In particular, we will show that PIA is more significant in reorienting the angular distributions of the ions toward the laser polarization direction.

A simple scheme for measuring the alignment-dependent ionization rates of arbitrary molecules has been proposed and exploited recently by Alnaser *et al* (2004) and Voss *et al* (2004). By choosing a suitable range of laser peak intensities and pulse durations in the so-called non-sequential double ionization regime (Walker *et al* 1994), they determined the kinetic energy release (KER) and the angular distributions of the product atomic ions in coincidence, in the double ionization of molecules by lasers over the 4π angles using the COLTRIM apparatus (Ullrich *et al* 1997, Dörner *et al* 2000). In the non-sequential double ionization region, which has been well-studied for D_2 molecules (Niikura *et al* 2003, Alnaser *et al* 2003, Tong *et al* 2003a, 2003b), double ionization proceeds through three successive steps. For O_2 molecules, for example, the molecule is first ionized at its equilibrium distance. Within an optical cycle or two, the tunnelled electron returns to excite O_2^+ to an excited electronic state which is easily further ionized by the laser field. The resulting O_2^{2+} then breaks up by Coulomb explosion, releasing kinetic energies characteristic of the specific O_2^{2+} electronic states and the internuclear separation R_o . For the electron impact excitation process, we can reasonably assume, which will be further justified later, that the process is nearly isotropic. The last step, laser ionization from the excited electronic states, is also isotropic since the laser intensity is above the saturation intensity for the excited states. Therefore, it was expected that the measured angular distribution of the coincidental ions reflects the alignment of the molecules at the instant of ionization weighted by the alignment-dependent ionization probabilities. By using a long pulse (35 fs) and a short pulse (8 fs) at laser intensities ranging from 1 to $10I_0$ ($I_0 = 10^{14} \text{ W cm}^{-2}$) (Voss *et al* 2004), the measured angular distributions, to our surprise, were found to peak much further toward the laser polarization direction than the predicted ones (Zhao *et al* 2003), despite the fact that the dynamic alignment of molecules before ionization has been properly included. The discrepancy prompted us to identify the additional post-ionization alignment effect.

When a homonuclear diatomic molecule is placed in a laser field, due to the anisotropic dipole polarizability, each molecule will receive a torque from the laser field. We can first estimate the PIA effect classically. Consider a Gaussian laser pulse with pulse duration τ (FWHM), we can calculate the angular momentum received by each molecule from the laser pulse,

$$L = \frac{1}{2}(\alpha_{\parallel} - \alpha_{\perp}) \sin(2\theta_0) \int_{-\infty}^{\infty} E^2(t) dt = \sqrt{\frac{\pi}{\ln 2}} \frac{I\tau}{8} (\alpha_{\parallel} - \alpha_{\perp}) \sin(2\theta_0) \quad (1)$$

where θ_0 is the initial angle of the molecule. Here α_{\parallel} and α_{\perp} are the parallel and perpendicular components of the polarizability with respect to the molecular axis, respectively, and $E(t)$ is the time-dependent laser field strength. Suppose at $t = 0$ (at the peak of the laser intensity) the molecule is first ionized and that we neglect the time interval between the first and the second ionizations. Thus the O_2^{2+} ion starts the breakup at $t = 0$. Due to the initial rotational angular momentum of the molecule at the second ionization, the axis between the two ions will rotate an additional angle until they are well separated. This angle is easily integrated to be

$$\delta\theta = \int_0^{\infty} \omega dt = \int_{R_0}^{\infty} \omega \frac{dR}{v(R)} = L \sqrt{\frac{2}{R_0\mu}}. \quad (2)$$

In deriving the above equation, we have used the following relations:

$$L = \mu R^2 \omega \quad (3)$$

$$v(R) = \frac{dR}{dt} = \sqrt{\frac{2}{\mu}(V(R_0) - V(R))}, \quad (4)$$

where μ is the reduced mass, R_0 is the equilibrium distance and $V(R)$ is the potential curve of the O_2^{2+} , which was approximated by $V(R) = 1/R$. For O_2 molecules, we get $\delta\theta = 0.066I\tau \sin(2\theta)$ in degrees. Here, I is the laser peak intensity in units of I_0 and τ is laser pulse duration in fs. Based on this simple classical model, the PIA effect will rotate the axis between the two ions by about 5° for a 35 fs laser pulse at $2.2I_0$. Note that this classical PIA is the consequence of the breakup of a rotating linear molecule. It is not very sensitive to whether the laser fields are present or not during the breakup process.

We can incorporate a quantum mechanical analysis of the PIA effect following the standard treatment of the rotational motion of a homonuclear molecule in a laser field. Here we approximate the diatomic molecule as a 'rigid' rotor (Seideman 1995, Ortigoso *et al* 1999, Cai *et al* 2001, Dion *et al* 2002). Each initial rotational state $\Phi_{JM}(\theta, \phi, -\infty) = Y_{JM}(\theta, \phi)$ of the molecule evolves in the laser field following the time-dependent Schrödinger equation (in atomic units $m = \hbar = e = 1$);

$$i \frac{\partial}{\partial t} \Phi_{JM}(\theta, \phi, t) = \left[B \hat{J}^2 + \frac{1}{2} \vec{E}(t) \cdot \vec{\alpha} \cdot \vec{E}(t) \right] \Phi_{JM}(\theta, \phi, t). \quad (5)$$

Here B is the rotational constant, $\vec{\alpha}$ is the polarizability (tensor) and $\vec{E}(t)$ is the time-dependent laser field. The solution of the above equation, for each initial rotation state Φ_{JM} , can be written as

$$\Phi_{JM}(\theta, \phi, t) = \sum_{J'} C_{J'}(JM, t) Y_{J'M}(\theta, \phi). \quad (6)$$

The time-dependent coefficients $\{C_{J'}(JM, t)\}$ can be obtained by solving equation (5) using the split-operator-method in the energy representation (Tong and Chu 1997). The time-dependent alignment distribution can be obtained as

$$\rho(\theta, t) = \sum_{JM} \omega_{JM} |\Phi_{JM}(\theta, \phi, t)|^2, \quad (7)$$

where ω_{JM} is the weight according to the Boltzmann distribution. For the homonuclear molecule considered here, nuclear spin statistics has to be properly accounted for as well. For diatomic molecules in a linearly polarized laser field, the angular distribution, or the alignment, does not depend on the azimuthal angle ϕ .

The two equations above allow the determination of the alignment of homonuclear diatomic molecules in a laser field at each instant. It describes the dynamic alignment of molecules in the laser field as well as the rotational revival of the enhanced alignment after the laser field has been turned off. The model assumes that the molecules remain in a ground state and no ionization happens. To incorporate post-ionization alignment effect into the formulation, we assume that the doubly charged molecular ions are created at $t = 0$ and $R = R_0$ where the Coulomb explosion gets started. As the doubly charged molecular ion breaks, the dipole polarizability changes with R , and the velocity of the atomic ions relative to each other increases, following equation (4). In the meanwhile, the moment of inertia of the 'rigid rotor' O_2^{2+} increases quadratically with R . The time dependence of B and the dipole polarizability $\bar{\alpha}$ can be entered into the time-dependent equation (5) through their variations with $R(t)$. Note that equation (5) still exhibits nontrivial time dependence even after the laser has been turned off. (This was used in the classical estimates, equations (2)–(4).) The time-dependent equations are integrated until the two atomic ions are far apart at which time the angular distributions of the ion fragments are calculated.

From the calculated rotational wavepacket, we can obtain the angular distribution $\rho(\theta, t)$ of the molecules as a function of time. Here we choose the laser polarization to be along the z -direction. The degree of alignment is often characterized by the alignment parameter defined by

$$S = \langle \cos^2 \theta(t) \rangle = \int \rho(\theta, t) \cos^2 \theta \, d\Omega. \quad (8)$$

We solve equation (5) for the following two cases. (a) Dynamic alignment calculation before ionization: both B and $\bar{\alpha}$ are constant during the whole time propagation. (b) Post ionization alignment calculation: we assume the second ionization occurs at peak laser intensity ($t = 0$) and the molecules will be further aligned as the doubly charged molecular ions are breaking up until $R \rightarrow \infty$. The propagation is carried out with R -dependent rotational constant B and R -dependent $\bar{\alpha}$. The time-dependent internuclear separation $R(t)$ is calculated from equation (4) where $V(R)$ is the potential curve from which the O_2^{2+} dissociates.

Since the diatomic molecule (or molecular ion) is treated as a linear rotor, the moment of inertia depends quadratically on R and thus the rotational constant B is inversely proportional to the square of R . For the dipole polarizability, the component perpendicular to the internuclear axis is not expected to change much as R is increased. The parallel component is expected to increase almost linearly with R . In fact, we have used the quantum chemistry code GAMESS to obtain the dipole polarizability for O_2^{2+} at a number of R . The perpendicular component is nearly constant and a value of 7.32 was used. For the parallel component, the R -dependence was found to be well fitted by $\alpha_{\parallel}(R) = 11.88 \times (1.0 + (R - R_0))$ where $R_0 = 2.28$ is the equilibrium distance of O_2 .

Note that PIA continues to be effective even after the laser field has been turned off. The time integration has to be carried out until large internuclear separation where the molecular ion is fragmented into a pair of atomic ions.

We first compare the relative importance of DA and PIA for O_2 molecules exposed to a 35 fs laser pulse with a peak intensity of $2.2I_0$, (see figure 1). The angular distribution or the alignment density of the molecules at $t = 0$ (at the peak of the laser pulse) is shown as a dashed curve (green). Clearly the distribution is still almost isotropic showing little DA of the molecules by the laser.

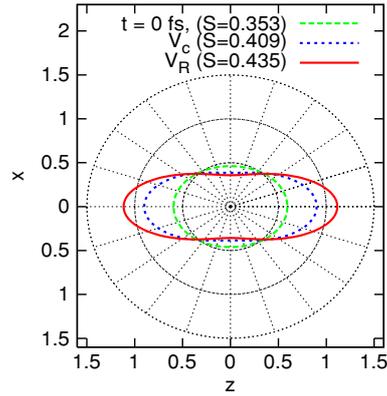


Figure 1. Alignment density distribution of O_2 molecules in a 35 fs pulsed laser field with laser intensity of $2.2I_0$. Dashed curve (green), dynamic alignment at $t = 0$. Dotted (blue) curves, including post-ionization alignment at $t \rightarrow \infty$, assuming the Coulomb potential; and solid (red) curve, using realistic molecular potential.

Assume that O_2^{2+} ions are created at $t = 0$ and the molecular ions start to break apart. If we assume that the internuclear potential is given by $V(R) = 1/R$, then the angular distribution of the atomic ions after the breakup is given by the dotted curve (blue). On the other hand, if we use the actual molecular potential curve of the O_2^{2+} from which the molecular ion breaks, the calculated angular distribution is much more forward peaked, as shown in the solid line (red). This clearly shows that PIA is much more important and is responsible for the forward angular distribution of the fragmentation products. In this calculation, we use the $O_2^{2+} \ ^3\Delta$ potential curve calculated by Larsson *et al* (1990). Even though we have assumed that O_2^{2+} was created at $t = 0$, the actual breakup may happen around the peak of the laser intensity or may be delayed by a few femtoseconds. We have made simulations for breakup starting at $t = -5, 5, 10$ fs and found the final angular distribution not sensitive to the initial breakup time. Recall that PIA effect persists even after the laser pulse has been turned off. In figure 1 we also indicate the alignment parameter S for the three different angular distributions. Even though S does not change much, the angular distribution has actually changed substantially. This indicates that S as defined in equation (8) is not a sensitive measure of the angular distribution of the molecules.

We next combine the two alignment effects discussed above, together with the alignment-dependent tunnelling ionization rates, calculated using the MO-ADK theory (Tong *et al* 2002), to predict the expected angular distribution of the fragmented ions that have been measured by Alnaser *et al* (2004) and Voss *et al* (2004). For a peak laser intensity of $2.2I_0$, double ionization proceeds non-sequentially. Initially the O_2 molecule is first ionized by the laser from its outermost π_g orbital at its equilibrium distance, with an alignment-dependent MO-ADK ionization rate. To compare with experiments we need to account for the pulse length and volume effects. We proceed this by following Tong *et al* (2002). By assuming that the temporal and spatial profiles of the laser pulse are Gaussian, we calculated the alignment-dependent ionization signals $R(\theta) = R_{\pi_g}(\theta)$ from the MO-ADK rates. We also make the additional assumption that the electron impact excitation cross section in the laser field is isotropic since such cross sections are not available. We justify this partly by showing later that such an assumption can explain the experimental results for double ionizations by a short laser pulse where alignment of the molecules due to the laser is not important. Thus, within such a model,

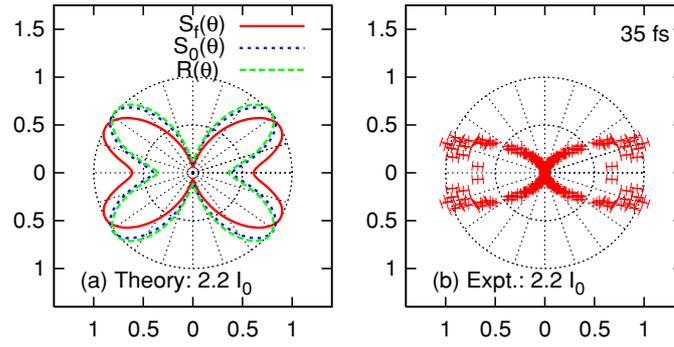


Figure 2. Comparison of the simulated angular distributions of the fragmented O^+ ions for 35 fs pulsed laser fields at $2.2I_0$. R is the alignment-dependent ionization signal; S_0 is the angular distribution neglecting PIA, and S_f is the same distribution including PIA.

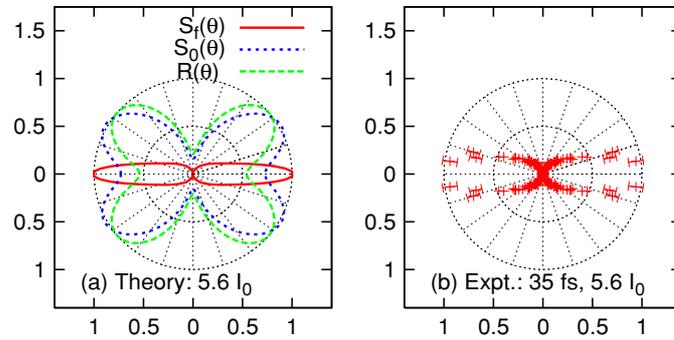


Figure 3. Comparison of the simulated angular distributions of fragmented O^+ ions with the measured ones for 35 fs pulsed laser fields at $5.6I_0$. R is the alignment-dependence ionization signal, S_0 is the angular distribution neglecting PIA and S_f is the same distribution including PIA.

if the PIA effect is neglected, the two O^+ ions would just fly apart at the alignment angle of the O_2 at which the first ionization occurs. In this case, the angular distribution of the two O^+ ions should be given by

$$S_0(\theta) \sim \rho(\theta, t=0)R(\theta). \quad (9)$$

If the PIA effect is included, then the theoretical prediction of the angular distribution is given by

$$S_f(\theta) \sim \rho(\theta, \infty)R(\theta). \quad (10)$$

In figure 2(a), we show the calculated theoretical angular distributions of the fragmented ions using the three models for the 35 fs pulses at the peak intensity of $2.2I_0$, by comparing $R(\theta)$, $S_0(\theta)$ and $S_f(\theta)$. Clearly there is little dynamic alignment up to $t = 0$. The major shift in the angular distribution toward the laser polarization direction comes from the PIA effect. Comparing with the experimental data of Voss *et al* (2004) shown in figure 2(b), the experimental data show evidence for the PIA effect but are more forward peaked than the theoretical calculations. We will come back to discuss the discrepancy later.

Figure 3 shows the theoretical calculations of the angular distributions for 35 fs pulses with a laser peak intensity of $5.6I_0$ and the comparison with experiment of Voss *et al*. Clearly the dynamic alignment at $t = 0$ is still not significant, but the PIA effect has a major influence

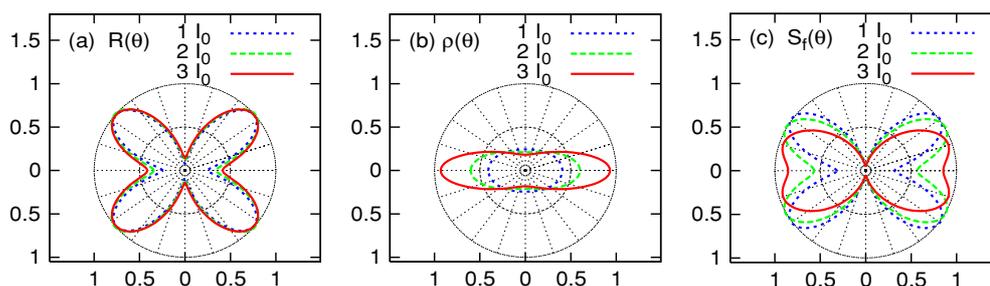


Figure 4. (a) Simulated alignment-dependent ionization signals for isotropic molecular oxygen, (b) alignment density distributions including DA and PIA and (c) final alignment-dependent ionization signal for 35 fs laser pulses.

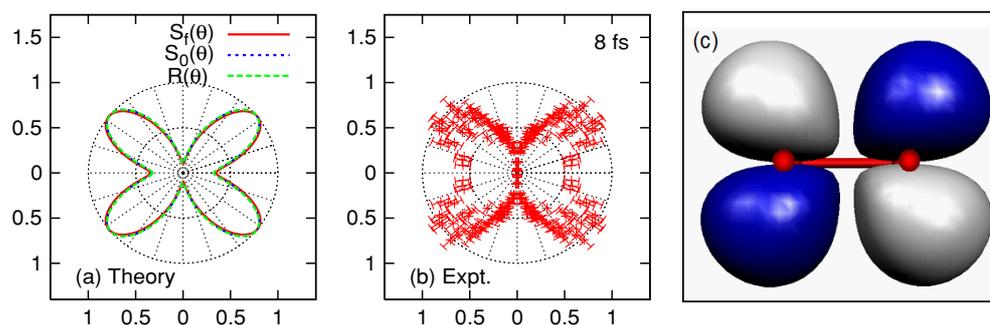


Figure 5. Ionization by an 8 fs laser pulse with peak laser intensity of $2I_0$. (a) Theoretical simulation shows neither dynamic alignment nor post-ionization alignment for such a short pulse. (b) Measured experimental angular distribution of the fragmented O^+ ions. (c) The angular density distribution of the outermost π_g molecular orbital of O_2 .

in shifting the angular distributions toward the forward direction. In fact, the peak of the predicted angular distribution of the fragmented ions is in the laser polarization direction, as a consequence of PIA effect. Comparing to the experimental data, the theoretical angular distributions are somewhat more forward peaked, but the qualitative agreement is good.

Both the theoretical and experimental data in figures 2 and 3 show that PIA effect is very important in aligning the fragmented ions toward the forward direction and the effect is very sensitive to the peak laser intensity. To illustrate such sensitivity, we show in figure 4 the ionization signals $R(\theta)$, the alignment density distribution $\rho(\theta, \infty)$ due to DA and PIA only, and the final predicted angular distributions $S_f(\theta)$ at three laser intensities of I_0 , $2I_0$ and $3I_0$. While the ionization rates do not change much at these three intensities, clearly PIA makes a significant change of $\rho(\theta, \infty)$ in going from $2I_0$ to $3I_0$. In fact, for $3I_0$, the calculated $S_f(\theta)$ shifts the peak of the angular distribution to a much smaller angle, closer to what the experiment shows in figure 2(b) for $2.2I_0$.

From the two examples above it is clear that for the 35 fs pulse the measured angular distributions of the fragmented ions are dominated by the PIA effect and provide limited information about the alignment dependence of the ionization rates directly. A much shorter laser pulse is needed if the measured angular distributions of the O^+ ions are to reveal the alignment-dependent ionization rates. Figure 5 shows the results obtained with an 8 fs laser pulse with peak intensity of $2I_0$. According to the theoretical calculations, for such a short pulse, there is little dynamic alignment, nor is there evidence of the PIA effect. Thus

the measured angular distributions of the fragmented O^+ ions should reveal the alignment-dependent ionization rates of O_2 molecules. Indeed, comparing with the experimental data, the measured angular distributions are in good agreement with the prediction from the MO-ADK theory. The comparison of theory and experiment shown in figure 5 is of particular interest. According to the MO-ADK theory, the alignment dependence of the ionization rates of a molecule is a direct measure of the angular distributions of the electronic density of the valence electrons of a molecule. In figure 5(c) the electronic charge cloud distributions of the outermost π_g orbital of O_2 calculated by GAMESS code (Schmidt *et al* 1993) are shown. The calculated electronic wavefunction is visualized by MOLEKEL program (Portmann and Lüthi 2000). Note that its angular shape resembles the calculated MO-ADK rates shown in figure 5(a). The latter in turn is in close resemblance with the measured angular distributions of the fragmented O^+ ionized by the 8 fs laser pulse. Thus for the first time the theoretical construct of molecular orbitals has been directly measured experimentally.

The good agreement between the simulation and experiment shown in figure 5 also support our model that there is no alignment dependence from the electron impact excitation and from the second ionization by the laser, in the non-sequential double ionization regime considered here. Such assumptions were made in the theoretical simulation of the spectra in figures 2 and 3.

We now return to discuss the discrepancy between theory and experiment in figure 2 at $2.2I_0$ and in figure 3 at $5.6I_0$. For the latter, the angular distribution is sharply forward peaked and the discrepancy is probably partly due to the failure in the experiment to measure the ionization rates in the forward direction. For the former, the major discrepancy probably can be attributed to the uncertainty in the experimental peak laser intensity which was estimated to be 30%. Based on the results in figure 4 it is clear that a higher laser intensity than $2.2I_0$ would easily account for the discrepancy. Note that it has been argued that a multiphoton process (Gibson *et al* 1997, Bandrauk and Ruel 1999) can contribute to the forward peaking of the fragmented ions as well. We have not been able to estimate this contribution quantitatively and thus its effect has not been included.

To summarize, we have shown quantitatively that post-ionization alignment effect plays a major role in determining the angular distributions of the fragmented ions in the double ionization of molecules by short intense laser pulses. By using a short 8 fs pulse at intensities in the so-called non-sequential double ionization regime, we showed that the post-ionization alignment effect is negligible and the experimental angular distributions of the fragmented ions provide a direct measure of the alignment-dependent ionization rates of a diatomic molecule. The latter, according to the MO-ADK theory of Tong *et al* (2002), is a direct reflection of the angular distribution of the molecular orbital of the outermost electron. Thus for the first time, it is possible to directly observe the electron cloud distribution of a molecular orbital experimentally without the complications from the final states.

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