ON ORIENTATION DEPENDENCE OF $N_2$ IONIZATION: THE VELOCITY GAUGE VERSION OF MOLECULAR STRONG-FIELD APPROXIMATION

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We reply to the Comment of T. K. Kjeldsen and L. B. Madsen [e-print arXiv:physics/0508213] and acknowledge their criticism related to imperfect composition of our model $3\sigma_g$ molecular state [V. I. Usachenko and S.-I. Chu, Phys. Rev. A 71, 063410 (2005)]. However, we cannot agree with the authors suggesting this critique also as an irrefutable argument and/or evidence in support and justification of their opposite and incorrect (viz. inconsistent with relevant experiment) orientation dependence of $N_2$ ionization rate calculated within the velocity gauge version of molecular strong-field approximation. We disclose the actual reason of the contradiction and demonstrate that appropriately composed $3\sigma_g$ state (modified according to the Comment’s critique) rather confirms the alternative earlier calculation [A. Jaron-Becker, A. Becker and F. H. M. Faisal, Phys. Rev. A 69, 023410 (2004)] suggesting correct orientation dependence of $N_2$ ionization, contrary to respective results of the Comment’s authors applying the same approach and procedure of $3\sigma_g$ composition.

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1. INTRODUCTION.

The current state-of-the-art in strong-field molecular ionization theory seems to be still far away from a sufficiently clear insight due to numerous controversial results obtained within different approaches and methods. As an appropriate illustration to mention here is the problem of strong-field ionization in laser-irradiated \( N_2 \) molecule, which recently received a special consideration within both the velocity-gauge (VG) [1-5] and the length-gauge (LG) [3, 6] formulations of conventional strong-field approximation (SFA) [7]. In particular, the orientation behavior of \( N_2 \) ionization rates was found in [3] to be quite different if calculated in the VG version versus the LG version of the molecular SFA approach and/or molecular tunneling theory [8]. Based on the same self-consistent Hartree-Fock (HF) numerical procedure as previously proposed in [1, 2] for the \( 3\sigma_g \) highest-occupied molecular orbital (HOMO) composition, the VG-SFA results [3] suggested a predominant \( N_2 \) ionization when the molecular axis is perpendicular to the incident laser field polarization (see also Fig. 2 in the Comment [4]). Thus, in contrast to their LG-VGA results [3], the VG-SFA results of the Comment’s authors suggest a counterintuitive behavior for \( N_2 \) ionization, which contradict to both relevant experiment [9] and alternative calculation [2]. Even though the Comment [4] applies in fact the same VG-SFA approach and numerical (HF-based) procedure of \( 3\sigma_g \) composition as earlier proposed in [1, 2], the Comment’s authors carefully avoid of quoting the opposite results of Ref.[2] and/or clarifying the reason of the contradiction. They prefer instead to criticize our imperfect composition of model \( 3\sigma_g \) molecular state [5] as inappropriately symmetrized and oversimplified. Moreover, they even suggest their criticism (being physically correct and acceptable as such) as an indispensable explanatory reason of the contradiction. In order to demonstrate the invalidity of such an interpretation (as well as the related Comment’s statements), we have to identify the actual reason of the contradiction by means of applying a more accurate molecular \( 3\sigma_g \) state appropriately modified according to the Comment’s critique.

2. THE LCAO-MO METHOD AND ASSOCIATED MOLECULAR WAVEFUNCTIONS OF \( 3\sigma_g \) STATE IN COORDINATE AND MOMENTUM SPACE.

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According to the standard linear combination of atomic orbitals (LCAO) and molecular orbitals (MO) method, the molecular orbitals are the mathematical constructs used to model the multi-electron molecular valence shells (each of a fixed discrete binding energy \( \varepsilon_{0}^{(n)} = -t_{p}^{(n)} \) and respective number \( N_{e}^{(n)} \) of identical electrons, similar to atomic orbitals in atom). For 3\( \sigma_{g} \) HOMO of gerade and bonding symmetry in \( N_{2} \) molecule under consideration, the corresponding approximate two-centered single-electron molecular wavefunction can be composed, for example, as an appropriate superposition of scaled hydrogen-like 2\( p_{z} \) and 1\( s \) (or 2\( s \)) atomic orbitals:

\[
\Phi_{(2p)}3\sigma_{g} (\mathbf{r}; R_0) = \frac{N_{e}^{(3\sigma_{g})}}{\sqrt{2 \left[ 1 - S_{2p_{z}}^{(3\sigma_{g})} (R_0) \right]}} \left[ \phi_{2p_{z}}^{(3\sigma_{g})} (\mathbf{r} - \mathbf{R}_0/2) - \phi_{2p_{z}}^{(3\sigma_{g})} (\mathbf{r} + \mathbf{R}_0/2) \right] \tag{1}
\]

\[
\Phi_{(1s)}3\sigma_{g} (\mathbf{r}; R_0) = \frac{N_{e}^{(3\sigma_{g})}}{\sqrt{2 \left[ 1 + S_{1s}^{(3\sigma_{g})} (R_0) \right]}} \left[ \phi_{1s}^{(3\sigma_{g})} (\mathbf{r} - \mathbf{R}_0/2) + \phi_{1s}^{(3\sigma_{g})} (\mathbf{r} + \mathbf{R}_0/2) \right] \tag{2}
\]

which are centered on each of the atomic cores and thus separated by internuclear distance \( R_0 \). Here \( N_{e}^{(3\sigma_{g})} = 2 \), whereas

\[
S_{j}^{(n)} (R_0) = \int d\mathbf{r} \phi_{j}^{(n)} (\mathbf{r} + \mathbf{R}_0/2) \phi_{j}^{(n)} (\mathbf{r} - \mathbf{R}_0/2) \tag{3}
\]

denotes the respective atomic orbital overlap integral.

Note that presently the ”−” combination of 2\( p_{z} \) states in Eq.(1) is used, which does provide the even spatial parity of the (2\( p \)) 3\( \sigma_{g} \) HOMO. Thus, even though the ”+” combination of scaled hydrogen-like 2\( p_{z} \) atomic orbitals alone earlier used in the literature [5, 6] to reproduce 3\( \sigma_{g} \) HOMO in \( N_{2} \) proved to be a surprisingly well working in explaining relevant experiment [10, 11], we have to mention here the correct Comment’s critical remark that such combination doesn’t provide the even (gerade) spatial parity and bonding symmetry of the (2\( p \)) 3\( \sigma_{g} \) MO. Moreover, according to the MO-SFA model [1], the oversimplified composition (1) would result in destructive intramolecular interference (see also [1-2, 5]) and accordingly high suppression in \( N_{2} \) ionization (contrary to the experiment [10, 11], which shows no suppression). To eliminate the mentioned deficiency, in our present consideration a more accurate composition of the 3\( \sigma_{g} \) HOMO is applied taking into account some admixture of a comparable contribution from atomic s-states mostly required to provide a good agreement with experiment (see also Sec.3 for details). Thus, in compliance with the second Comment’s critical remark, the 3\( \sigma_{g} \) HOMO is to be further approximated by the coherent superposition of few different MOs corresponding to separate contributions from atomic states of a specified orbital symmetry (viz., the scaled hydrogen-like 1\( s \), 2\( s \) and 2\( p_{z} \) orbitals) under consideration:

\[
\Phi_{(1s2s2p)}3\sigma_{g} (\mathbf{r}; R_0) = A_{1s}\Phi_{(1s)}3\sigma_{g} (\mathbf{r}; R_0) + A_{2s}\Phi_{(2s)}3\sigma_{g} (\mathbf{r}; R_0) + A_{2p}\Phi_{(2p)}3\sigma_{g} (\mathbf{r}; R_0) \tag{4}
\]

with the weight coefficients \( A_{j} \) \((j = 1, 2, 3)\) being the relative contributions \(|A_{j}| \leq 1\) from respective atomic states and considered as variational parameters to be found from the equation for minimum of respective molecular binding energy, the value of which is put to be equal to the experimental value.
The model single-electron molecular wavefunctions (1)-(2) allow for analytical representation of the Fourier transform (or respective molecular state corresponding to a definite value of final photoelectron momentum \( p \) in momentum space) for each of one-electron two-centered single molecular orbitals contributing to the \( 3\sigma_g \) HOMO in \( N_2 \) under consideration:

\[
\Phi_{(2p)3\sigma_g}(p_N, R_0) = -\sqrt{N_e(3\sigma_g)} C (\kappa_n) \frac{2^5\kappa_n^{7/2} p_N \cos (\theta_p)}{\pi \sqrt{2 (p_N^2 + \kappa_n^2)^3}} \sin \left[ \frac{(p_N \cdot R_0)}{2} \right] \sqrt{2 \left[ 1 - S_{2p_z} (R_0) \right]} \tag{5}
\]

\[
\Phi_{(2s)3\sigma_g}(p_N, R_0) = \sqrt{N_e(3\sigma_g)} C (\kappa_n) \frac{2^4\kappa_n^{5/2} (p_n^2 - \kappa_n^2)}{\pi \sqrt{2 (p_N^2 + \kappa_n^2)^3}} \cos \left[ \frac{(p_N \cdot R_0)}{2} \right] \sqrt{2 \left[ 1 + S_{2s} (R_0) \right]} \tag{6}
\]

\[
\Phi_{(1s)3\sigma_g}(p_N, R_0) = \sqrt{N_e(3\sigma_g)} C (\kappa_n) \frac{2^{5/2}\kappa_n^{5/2}}{\pi (p_N^2 + \kappa_n^2)^2} \cos \left[ \frac{(p_N \cdot R_0)}{2} \right] \sqrt{2 \left[ 1 + S_{1s} (R_0) \right]} \tag{7}
\]

with the polar angle \( \theta_p \) of photoelectron emission relative to internuclear molecular axis \( \cos (\theta_p) = (p \cdot R_0)/pR_0 \). Here \( \varepsilon_0^{(n)} = -\kappa_n^2/2 = -I_p^{(n)} = -\left( Z_j^{(n)} / a_j \right)^2/2 \) is the binding energy of \( n \)-th initial molecular discrete state under consideration (viz., \( I_p^{(3\sigma_g)} (N_2) \approx 15.58 \text{ eV} \) and \( Z_j^{(n)} \) is the effective charge corresponding to "effective" long-range Coulomb model binding potential of respective residual molecular ion, while \( a_j = j a_0 \) is \( j \)-th Bohr orbital radius of respective contributing atomic orbital. The analytical expressions for respective atomic orbital overlap integrals (3) are presented in [5], moreover, the Coulomb-Volkov correction factor \( C (\kappa_n) = \left( 2\kappa_n I_p^{(n)} / E \right)^{\kappa_n^{-1}} \) is also introduced to matrix elements (5)-(7) to incorporate the long-range Coulomb electron-molecular ion interaction in the final continuum states due to the VG version of SFA applied [1].

3. DISCUSSION.

Fig.1 demonstrates the angular dependence of molecular wavefunction (4) in momentum space presented for different \( 3\sigma_g \) compositions, either with or without taking a comparable contribution from atomic \( s \)-states into account. Let’s note first that the angular dependence corresponding to contribution from \( 2p_z \) states alone (Fig.1a) is considerably prolate along the internuclear axis since the \( 2p_z \) states are predominantly ionized along the internuclear axis owing to the presence of the geometrical factor \( \cos \theta_p \) in Eq.(5) (cf. also with Eq.(20) and respective Fig.1a in [5]). This, particularly, illustrates that the spatial (gerade or ungerade) symmetry of respective \( (2p) \) \( 3\sigma_g \) MO doesn’t essentially affect the angular dependence (viz., nodal plane along the molecular axis) in momentum space. Thus, irrespectively of the sign in the combination of \( 2p_z \) atomic states, the respective \( (2p) \) \( 3\sigma_g \) MO suggests a predominant photoelectron emission along the internuclear axis in momentum space. The sign in the combination mostly alters the absolute value of local maxima in respective angular dependence due to different (sine or cosine) trigonometric factor arising from intramolecular interference of contributions of ionization from two separate atomic centers [1]. In particular, the \( \sin \left[ \frac{(p_N \cdot R_0)}{2} \right] \) factor in Eq.(5) corresponding to the "−" sign results in destructive intramolecular interference and a
suppressed molecular ionization (e.g. in $O_2$ molecule relative to $Xe$ atom, see also [1, 2, 5], for details).

Accordingly, if the currently used "−" combination of $2p_z$ states is supposed to contribute to the $3\sigma_g$ MO, then a coherent contribution from other ($1s$ and/or $2s$) atomic states should be also taken into equally detailed consideration as described by Eq.(4). The respective Figures 1c-1d (versus Fig.1a) demonstrate that the contribution from $s$-states to the $(1s2p)\ 3\sigma_g$ and $(1s2s2p)\ 3\sigma_g$ MO dominates in momentum space at low photoelectron momenta, in a qualitative accordance with stated in the Comment. Unfortunately, the Reply’s restrictions don’t allow us to provide the reader with an extensive and sufficiently detailed information about the respective molecular ionization rates, it will be given elsewhere in our further related publications. So far we only have to stress that the corresponding relative (weight) coefficients $A_j$ at $1s$, $2s$ and $2p_z$ states under consideration proved to have comparable values, although not necessarily of the same sign. Namely, the $3\sigma_g$ compositions with $A_{1s}$ and $A_{2p}$ (as well as $A_{1s}$ and $A_{2s}$) of opposite signs only proved to provide no suppression observed in experiment [10, 11] for $N_2$ ionization as compared to ionization of counterpart atomic $Ar$ of nearly identical ionization potential $I_p^{(Ar)} \approx 15.75$ eV. In particular, the total ionization rates calculated for $A_{1s}$ and $A_{2p}$ of opposite signs in $(1s2p)\ 3\sigma_g$ and $(1s2s2p)\ 3\sigma_g$ MO proved to demonstrate no suppression (and even some enhancement) in $N_2$ ionization relative to $Ar$ well consistent with relevant experiment [10, 11]. Our Figs.1c-1d also suggest that $(1s2p)\ 3\sigma_g$ and $(1s2s2p)\ 3\sigma_g$ MO corresponding to the coefficients $A_{1s}$ and $A_{2p}$ of opposite signs are always noticeably prolate in momentum space along the internuclear axis, although to a considerably less extent as compared to the $(2p)\ 3\sigma_g$ composed of pure $2p_z$ states alone.

Anyhow, our Figs.1c-1d clearly illustrate that a more accurate consideration doesn’t necessarily result in the $3\sigma_g$ HOMO to predominate in momentum space along the direction perpendicular to the internuclear axis (as suggested, e.g., by Fig.4e of the first reference in [3] and/or Fig.1f in the Comment [4]). Since the generalized Bessel function are commonly maximized when photoelectron momentum $p$ is parallel to the laser field polarization $e$, the ionization from $(1s2p)\ 3\sigma_g$ and $(1s2s2p)\ 3\sigma_g$ presented in our Fig.1 is expected to be predominant when the $N_2$ molecular axis is aligned along the laser field $(R_0||e)$. This difference thus invalidates the Comment’s statement that incorrect (viz., inconsistent with experiment) orientation dependence of $N_2$ ionization rate the authors also found in [3] is just explained by the VG version of molecular SFA they applied. Our Fig.2 well illustrates the above conclusion and displays the orientation dependence of total $N_2$ ionization rates (see Eq.(13) in [5]) corresponding to different $3\sigma_g$ compositions. As expected, the compositions of $3\sigma_g$ HOMO taking a comparable contribution from $p$-states into account always suggest a predominant ionization of $N_2$ for parallel orientation of internuclear axis relative to the laser field polarization. Whereas, the composition of $3\sigma_g$ accounting for a contribution from pure $s$-states alone is well seen to only give a predominant ionization of $N_2$ for perpendicular orientation of internuclear axis as suggested in the Comment. Such a behavior may thus arise only due to the contributing $1s$ or $2s$ states, which are spherically symmetric and equally well ionized along any spatial direction. Therefore, the form of respective nodal plane relative to the molecular axis suggested by Eqs.(6) and (7) is solely determined by the cosine trigonometric
factor $\cos \left( \frac{\mathbf{p}_N \cdot \mathbf{R}_0}{2} \right)$, which alone suggests the angular dependence to be oblate along the internuclear axis in momentum space (Fig.1b). The resulting orientation dependence with maximum ionization for the perpendicular orientation of molecular axis is thus quite naturally expected for ionization of $H_2$ and $H_2^+$ (with a single $(1s) 1\sigma_g$ MO composed of pure $s$-states). For $H_2$, such orientation dependence was recently revealed [12] using a tunnelling model with a Heitler-London type valence bond wavefunction.

To summarize, our argumentation based on a more accurate consideration of $3\sigma_g$ HOMO (4) is also qualitatively well consistent with alternative calculation [2] using the same VG-SFA approach as applied by the Comment’s authors [3, 4]. Meantime, we also have to stress that opposite orientation dependence of $N_2$ ionization presented in Fig.2 of the Comment [4] is based on the same numerical HF-based procedure of $3\sigma_g$ HOMO composition as applied in [2] under precisely the same situation (cf. with respective Fig.7b in [2]). Therefore, we believe that orientation dependence of $N_2$ ionization rate presented in our Fig.2 is correct and its consistence with relevant experiment [9] not just an accidental. By means of direct comparison of our Fig.1c (and/or Fig.1d) with respective Fig.4e presented in [3], one can identify that opposite behavior of $N_2$ ionization suggested in [4] (as well as earlier in [3]) stems from a different angular dependence of $3\sigma_g$ molecular state in momentum space. Our Fig.1 also suggests that such difference may only arise owing to relative contribution from $s$-states, which seems to be somehow overestimated (compared to $p$-states) within the particular $(12s7p)/(6s4p)$ basis chosen in the Comment for numerical HF-based procedure of $3\sigma_g$ composition in coordinate space. This, particularly, becomes especially evident from Fig.3 displaying the correct orbital shape for the $3\sigma_g$ wavefunction in coordinate space presented in Ref.[13] (see Fig.12 therein), which obviously differs by a remarkably prominent domination of $p$-states from the resulting $3\sigma_g$ state of substantially different shape and detailed structure suggested in [4] (cf. respective Fig.1c therein). Meantime, the numerical procedure applied in [3, 4] for $3\sigma_g$ composition doesn’t seem to be sufficiently transparent with respect to the extent of such relative contribution, which is thus hardly amenable to reliable analysis and/or available for direct comparison [14]. Note also that a composition overestimating the relative contribution from $s$-states in coordinate space is not suitable to $3\sigma_g$ state in $N_2$, which is known to have a different and more complex structure well dominated by $p$-states [15, 16].

Finally, we also have to clarify the main message of a different (although, somewhat related) research [17] the Comment’s authors quite inappropriately and very ambiguously invoked in support of their related statement suggesting the LG version to be superior to the VG version of the SFA under discussion. Recall that the cited similar conclusion of Ref.[17] (see also Fig.1 therein to make certain) applies solely to ionization of atomic states of only odd spatial parity, such as $p$-states, which are not symmetric with respect to the inversion of all electron coordinates to opposite direction. Whereas, for the molecular $3\sigma_g$ state (which is known as surely gerade, i.e. of even spatial parity), the VG and LG versions of SFA, according to [17], are expected to give the same results, contrary to respective VG-SFA findings of Ref.[3] and the context of main related statement of the Comment [4].

4. CONCLUDING REMARKS.
The criticism of the Comment [4] related to our previous imperfect composition of the $3\sigma_g$ molecular wavefunction [5] was assessed to be physically correct as such, however not affecting essentially our related final results and conclusions based on the velocity gauge (VG) version of molecular strong-field approximation (SFA). In particular, when applying an appropriately modified $3\sigma_g$ wavefunction, we demonstrate that orientation dependence of $N_2$ ionization is to be correct (viz., consistent with experiment) and thus the Comment’s critique cannot be accepted as an irrefutable argument and indispensable evidence in support of the authors’ opposite respective VG-SFA findings [3, 4]. This, particularly, invalidates the Comment’s main statement about unavailability of the SFA in its VG formulation for adequate description of molecular strong-field ionization when an accurate molecular orbital is applied. Our conclusion is also well consistent with recent Erratum [18] suggesting in fact analogous corrections to analytical expressions for molecular ionization rate previously numerically calculated in [1, 2] for diatomic molecule having a HOMO of gerade symmetry. Those corrections were also confirmed in [18] as not affecting their correct final results and conclusions discussed elsewhere earlier including the interference suppression of molecular ionization and orientation dependence of $N_2$ ionization [1, 2], which proved to be quite opposite to respective VG-SFA result of the Comment applying the same approach and pure numerical HF-based procedure of $3\sigma_g$ composition.

5. ACKNOWLEDGMENTS.

This work was supported by Center of Sciences and Technology of Uzbekistan (Project No. Φ-2.1.44). The support from DAAD (Deutscher Akademischer Austauschdienst) and B-Division of Max-Born-Institute of Nonlinear Optics and Short-Pulse Laser Spectroscopy (Berlin, Germany) is also gratefully acknowledged.

The research described in this presentation was made also possible in part by financial support from the U.S. Civilian Research and Development Foundation (CRDF) for the Independent States of the Former Soviet Union.

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FIGURE CAPTIONS

1. Fig.1 (Color online). The squared module $|\Phi_{3\sigma_g}(\mathbf{p}_N, \mathbf{R}_0)|^2$ of $3\sigma_g$ HOMO wavefunction in momentum space (the Fourier transform) plotted vs the angle $\theta_p$ of photoelectron emission (relative to the internuclear molecular axis lined up in the horizontal direction) and calculated for four different $3\sigma_g$ compositions: (a) $(1s)\ 3\sigma_g$ MO composed from 1s states alone, (b) $(2p)\ 3\sigma_g$ MO composed from $2p_z$ states alone, (c) $(1s2p)\ 3\sigma_g$ MO composed from 1s and $2p_z$ states, (d) $(1s2s2p)\ 3\sigma_g$ MO composed from 1s, 2s and $2p_z$ states. These angular dependencies are all presented for photoelectrons only emitted along the polarization of Ti:sapphire incident laser field of the wavelength $\lambda = 800\ nm$ ($\hbar \omega = 1.55\ eV$) and fixed intensity $I = 2 \cdot 10^{14}\ W/cm^2$ due to absorption of different number $N$ of laser photons beginning from minimum one $N_0 = 18$ required for ionization of $N_2$.

2. Fig.2 (Color online). The total $N_2$ ionization rates corresponding to $3\sigma_g$ compositions presented in Fig.1 and plotted vs the angle $\Theta$ of the internuclear axis orientation relative to the polarization of Ti:sapphire incident laser field of the wavelength $\lambda = 800\ nm$ ($\hbar \omega = 1.55\ eV$) and fixed intensity $I = 2 \cdot 10^{14}\ W/cm^2$. The ionization rates are all normalized to respective ones at the angle $\Theta = 0$ and presented for four different $3\sigma_g$ compositions: (1s)$\ 3\sigma_g$ MO composed from 1s states alone (solid line); (2p)$\ 3\sigma_g$ MO composed from $2p_z$ states alone (dashed-dotted line); (1s2p)$\ 3\sigma_g$ MO composed from 1s and $2p_z$ states (short dashed line), (1s2s2p)$\ 3\sigma_g$ MO composed from 1s, 2s and $2p_z$ states (long dashed line). This figure is to be compared with Fig.2a of the Comment and Fig.2a of relevant experiment [9].

3. Fig.3 (Color online). The orbital shape of the correct $3\sigma_g$ HOMO in $N_2$ molecule imaged by recording the high harmonic spectra and presented in [13] (see Fig.12 therein and related explanation). Note that this is not the orbital density, but a wavefunction to within a global phase. This figure is to be compared with respective Fig.1c of the Comment displaying the alternative $3\sigma_g$ HOMO numerically composed within particular $(12s7p) /[6s4p]$ basis set.
Fig. 1

(a) The $(2p)^3\sigma_g$ molecular orbital in momentum space (a. u.)

(b) The $(1s)^3\sigma_g$ molecular orbital in momentum space (a. u.)

(c) The $(1s2p)^3\sigma_g$ molecular orbital in momentum space (a. u.)

(d) The $(1s2s2p)^3\sigma_g$ molecular orbital in momentum space (a. u.)

$\theta_p$, Angle of Photoelectron Emission (degrees)
Fig. 2

Theta, Angle of Molecular Axis Orientation (degrees)
This figure "3SiG-Fig3.jpg" is available in "jpg" format from:

http://arxiv.org/ps/physics/0604068v1