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Role of high-lying electronic states on the angular distribution of photofragments in multiphoton dissociation of D_2^+ by intense laser fields

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We have theoretically investigated the role of high lying electronic states $2p\pi_u$ and $3d\sigma_g$ on multiphoton dissociation of D_2^+ from initial vibrational-rotational levels $v_i=4,5$ and $J_i=0,1$ by linearly polarized light with wavelength 400 nm and intensity 3×10^{13} W/cm^2. We have found that for initial level $v_i=4$ and $J_i=1$, a considerable number of photofragments is ejected perpendicular to the polarization direction of the laser field. It is shown that the perpendicular dissociation is due to the (net) four-photon absorption of the ground $1s\sigma_g$ state to the $3d\sigma_g$ state through the intermediate five-photon coupling of the $1s\sigma_g$ and $2p\pi_u$ states at intense laser field with high frequency. Our computed value of the emission of D^+ ions in the perpendicular direction to that in the parallel direction of the laser polarization reasonably agrees with the recent experimental result of Chin and co-workers at the same laser parameters [Phys. Rev. A **62**, 042708 (2000)].

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I. INTRODUCTION

For the last decade, experimental [1,2] and theoretical [3–5] investigations of the multiphoton dissociation of the simplest diatomic molecules, H_2^+, D_2^+, and HD^+, have provided us with a lot of insight about the behavior of molecules in intense laser fields. In particular, the study of angular distribution of dissociation fragments has posed many interesting questions about the intense field dynamics of molecules. The framework of the theoretical explanation involved only the two lowest electronic states in the multiphoton dissociation process. Until very recently, the experimental data over the available range of intensities and frequencies tended to confirm the theoretical picture.

The experimental work started about a decade ago with the observation of photofragments resulting from dissociation of H_2^+ molecules by intense linearly polarized laser fields of wavelength 532 nm by Bucksbaum and co-workers [1]. The intensities used were 5×10^{13} and 1×10^{14} W/cm^2. They found that the H^+ ions were emitted in a narrow distribution along the laser field polarization direction. The dissociation involved in the experiment certainly occurred from an unknown mixture of vibrational-rotational states of H_2^+. Recently, Chandler and co-workers [2] performed novel experimental investigations on the angular distribution of fragments produced from dissociation of D_2^+ by absorption of one, two, or three photons of wavelength 532 nm for peak intensities 1.5×10^{13} and 5×10^{13} W/cm^2. The branching ratios for different photon absorptions obtained by them confirmed the pattern previously predicted on theoretical considerations [3]. Mainly low initial vibrational levels $v_i\leqslant3$ of D_2^+ were expected to be present in their experiment. They observed that for linearly polarized light, the D^+ ions resulting from dissociation of D_2^+ were ejected in a narrow cone about the polarization axis of the laser field.

From previous experiments on molecules with intense picosecond laser pulses at 532 nm [1,2,6], it was well known that linearly polarized fields can result in spatial alignment of the internuclear axis along the polarization direction. For long pulses (with time duration of a few picosecond), such an alignment is dynamic and results from the realignment of the molecule along the field direction by the superposition of field-free molecular rotational states through repeated absorption and stimulated emission of photons. For a sufficiently long pulse, the molecule can be, very generally, shown to be in pendular states [6], which are states executing angular oscillations about the polarization direction. The signature of such pendular states was also obtained experimentally [6]. For very short pulses, however, the molecular rotational states may not be thoroughly mixed and the dissociation dynamics would more closely reflect the effects of geometric alignment.

It is interesting that, because of its ease of production, the 532 nm laser light was generally used in these experiments. Also, in the case of hydrogenic molecule, quite low vibrational levels were excited and they were not properly resolved, and the initial rotational level could not be ascertained. The angular distribution obtained in the experiments [1,2] did indeed confirm the theoretical expectations according to the conventional two-electronic-state approach. Detailed quantal calculation was made by Atabek and coworkers [4] on energy-resolved angular distribution of photofragments of H_2^+ from initial vibrational levels $v_i=2,4,5$ and rotational level $J_i=1$, with linearly polarized pulses of wavelength 532 nm and peak intensities 1×10^{13} and 5×10^{13} W/cm^2. With their two-electronic-state expansion, again H^+ fragments aligned with the laser polarization axis were obtained. This kind of alignment was due to the superposition of rotational states prior to dissociation. Very recently, we have studied the angular distribution of photofragments in multiphoton above threshold dissociation of HD^+ from the initial levels $v_i=0-2$ and $J_i=0$ by linearly and circularly polarized laser fields of a shorter wavelength

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*Electronic address: msscs@mahendra.iacs.res.in of 329.7 nm and for intensities 1×10^{12}, 5×10^{12}, and 1×10^{13} W/cm^2 [5]. Again, in two-electronic-state expansion, in case of linear polarization, the emission of photofragments was found to be confined to a narrow cone about the field polarization direction when sufficiently large number of rotational states was included in the expansion. Correspondingly, in the case of circular polarization, the photofragments were preferably emitted in a plane perpendicular to the propagation direction.

Intense field dissociation process is interpreted in terms of the motion of the nuclei on the field, dressed adiabatic surfaces and the nonadiabatic transitions in the region of the avoided crossings [3]. The positions of the avoided crossings depend on the frequency of the laser field, and the probability of following the adiabatic path will depend on the intensity. The dynamics, and hence the branching ratios and angular distributions are expected to be sensitive to the initial wave function. Experimental determination of the initial state resolved angular distribution as a function of frequency should ideally resolve these issues, but no such detailed experimental work exists. However, in a recent breakthrough, Chin and co-workers [7] have performed experimental investigations on dissociative ionization of D_2 molecules using linearly polarized intense laser pulses of wavelengths 400 nm and 800 nm with peak intensities 3×10^{13} W/cm^2 and 2×10^{14} W/cm^2, respectively. They measured the time-of-flight spectra of the ions and showed that during the dissociation of ${D_2}^+$ by the laser with the shorter wavelength of 400 nm, a significant number of D^+ ions was ejected perpendicular to the direction of polarization of the laser field. But for the dissociation of the ${D_2}^+$ molecular ions by the longer wavelength of 800 nm, the number of D^+ ions in the perpendicular direction to the field polarization was insignificant, as obtained by experimentalists [1,2] earlier for ${H_2}^+$, ${D_2}^+$ (notably at 532 nm) and predicted by theoretical three-dimensional (3D) calculations [4,5] for ${H_2}^+$, HD^+ on the basis of radiative coupling between the two lowest electronic states. In fact, the adiabatic passage through the one-photon crossing of the two lowest potential energy curves will be forbidden for perpendicular alignment to the laser field. The fact that photofragments were observed in a direction perpendicular to the laser polarization was interpreted in terms of the multiphoton radiative coupling of the ground $1s\sigma_g$ state with the high lying $2p\pi_u$ and $3d\sigma_g$ electronic states of ${D_2}^+$ [7]. The authors concluded that these highly excited electronic states play an essential role in multiphoton dissociation process from excited vibrational levels of ${D_2}^+$ at high frequencies and high intensities.

In the present work, we have theoretically investigated in detail the role of high lying electronic states $2p\pi_u$ and $3d\sigma_g$ on the multiphoton dissociation of ${D_2}^+$. We have paid particular attention to the modification of the angular distribution that such states may cause for photofragments with different kinetic energies for different initially excited vibrational-rotational levels. The theoretical calculations have been made in the context of the experiment discussed above, and for the 400 nm wavelength and 3×10^{13} W/cm^2 intensity as in the experiment [7]. We start from particular rotational-vibrational states and look for the stationary solutions with appropriate boundary conditions. This implies that the solutions would be particularly applicable for long pulses (in the picosecond regime) acting upon state selected molecules.

II. CALCULATIONS

We have used the time-independent close-coupling (CC) method [8]. The CC technique has been adopted in our earlier works [3,5], where the theory and justification of this approach have been discussed in detail. We have considered, in this work, four (five) electronic states of ${D_2}^+$ to be coupled by the radiation field (of $\lambda=400$ nm). The states are the ground $1s\sigma_g$ and the first excited $2p\sigma_u$ as well as the high lying $2p\pi_u^+$ $(2p\pi_u^-)$ and $3d\sigma_g$. Molecular rotation has been taken into account by including 96 channels with rotational quantum number $J=0$–7 and absorbed photon number $n=0$–7 in our basis set for linearly polarized light (which obeys the selection rule $\Delta M=0$). The initial vibrational-rotational levels considered here are $v_i=4,5$ and $J_i=0,1$. It may be noted here that because of the Σ-Π selection rule and the use of linearly polarized light, for initial $M_i=0$, transition to the $2p\pi_u^-$ state is forbidden. In this case, the total number of coupled channels considered reduced to 72 using the same angular momentum and photon number states mentioned above. The radiative couplings between all states have been considered in a truncated length gauge form of the interaction, as in earlier works [3,5,9].

In the present case, the distribution of photofragments for an initial rovibrational level v_i, J_i is given by, following Balint-Kurti and Shapiro [10],

$$P_i(\theta,E_0,\Lambda,n)=\frac{k}{(2J_i+1)}\sum_{M_i}\left|\sum_{JM}(i)^J(2J+1)^{1/2}\times D^{J*}_{M\Lambda}(\varphi,\theta,0)T_{JM\Lambda}(E_0,n|i)\right|^2, \qquad (1)$$

where $T_{JM\Lambda}(E_0,n|i)$ is the bound-free transition amplitude [5] for n-photon absorption evaluated at the energy E_0 for which the highest peak of the overlap between the initial bound wave function and the free-state scattering wave function occurs. This corresponds to the dominant laser induced resonant state corresponding to the initial vibrational level in the $1s\sigma_g$ potential. Due to the mixing of the rotational levels, there may be multiple peaks at various total energies [3], signifying the mixed character of the laser induced resonances in the $1s\sigma_g$ state. Thus, we have generally calculated the distribution at the most prominent resonance. θ is the angle between the momentum of the dissociated fragments and the space fixed Z axis, i.e., the light polarization direction. $D^J_{M\Lambda}$ is the Wigner matrix element [11] and k is some specific constant. It may be noted that P_i is independent of the azimuthal angle φ for axially symmetric molecules.

The Born-Oppenheimer (BO) potentials for the $1s\sigma_g$ and $2p\sigma_u$ states are obtained from Carrington and Kennedy [12], while those for the $2p\pi_u$ and $3d\sigma_g$ states are taken from Murai and Takutsi [13]. The $1s\sigma_g\rightarrow2p\sigma_u$ transition dipole moments are given by Bates [14], while the $1s\sigma_g\rightarrow2p\pi_u$,

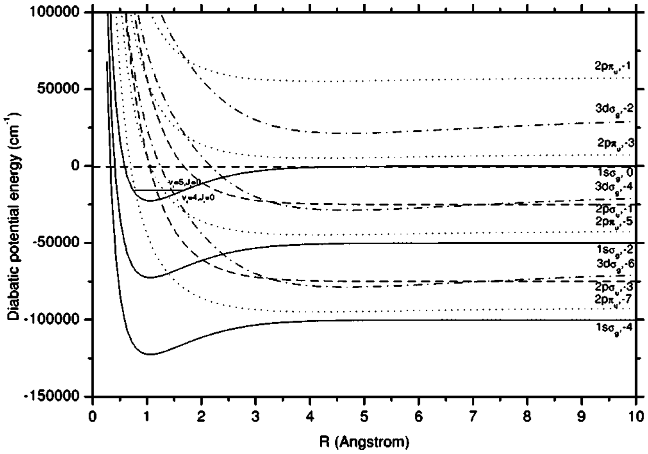

FIG. 1. Diabatic potential energy curves for four electronic states of $D_2{}^+$ with a number of 400 nm photon absorption. The unperturbed eigenenergies of the initial vibrational levels $v_i=4$ and 5 with $J_i=0$ are also indicated.

$2p\sigma_u \rightarrow 3d\sigma_g$, and $2p\pi_u \rightarrow 3d\sigma_g$ transition dipole moments are obtained from the extensive work of Ramekar and Peek [15].

III. RESULTS AND DISCUSSION

Some of the field plus molecule diabatic energy curves (i.e., the energy of the noninteracting field plus molecule system) have been plotted as functions of the internuclear distance *R* in Fig. 1 for the four relevant electronic states and a number of photon absorption of wavelength 400 nm. To keep the discussion simple, only the energy curves for a single rotational quantum number have been shown (for every field plus molecule state considered). Actually, the energy curves for each field plus electronic state should form a band with states of different *J* values, lying close together, one curve for each value of *J*, the rotational quantum number used. The diabatic curves cross at different internuclear distances, and at the crossing points the multiphoton couplings between different field plus electronic states will be effective in causing miltiphoton transitions between them. In Fig. 1, eigenenergies of the unperturbed $v_i=4$ and 5 levels for $J_i=0$ have also been indicated. We note that the region, in which the wave functions of these initial vibrational levels are appreciable, coincides with the region of the crossing of the ground $|1s\sigma_g,0\rangle$ state with the $|2p\pi_u,-5\rangle$ and $|2p\sigma_u,-1\rangle$ states. The $1s\sigma_g$-$2p\sigma_u$ coupling will cause bond softening and tunneling dissociation of the molecule by single-photon absorption as usual. However, at the intensities where five-photon coupling is effective, the molecules in the excited vibrational levels may directly absorb five photons to make transition to the $2p\pi_u$ state. This direct five-photon transition to the $2p\pi_u$ state was also suggested [7] as the reason of finding photofragments in a direction perpendicular to the laser polarization. Indeed, the perpendicular transition moment effective for the σ-π five-photon coupling near $R=1.8$ Å may cause interaction with photons even when the molecular axis is perpendicular to the polarization direction of light.

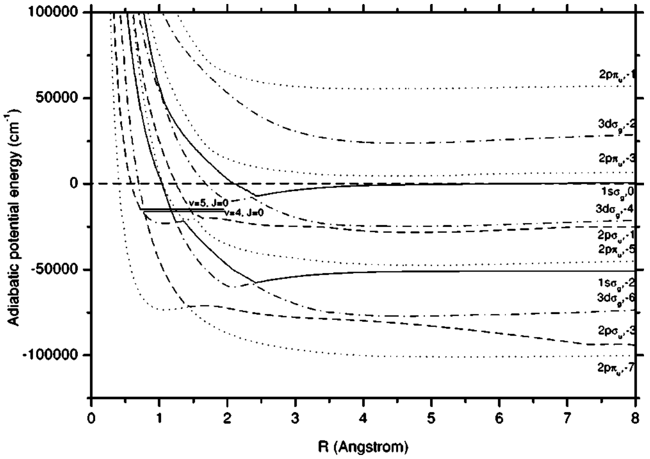

FIG. 2. Adiabatic potential energy curves for four electronic states of $D_2{}^+$ at laser wavelength 400 nm and intensity 3×10^{13} W/cm^2. The dressed energies corresponding to the initial field-free vibrational levels $v_i=4$ and 5 with $J_i=0$ are also shown.

Figure 2 shows a very simplified version of the adiabatic

TABLE I. The ratios $\mathfrak{R}$ of the D^+ ions ejected in the perpendicular direction ($\theta=90°$) of the laser polarization to the total D^+ ions emitted along the laser polarization direction ($\theta=0°$) are shown for initial levels $v_i=4,5$ and $J_i=0,1$ of $D_2{}^+$ at laser wavelength 400 nm and intensity 3×10^{13} W/cm^2. Also, the contributions to the angular distributions (P_i) at $\theta=0°$ and 90° from relevant final channels are given. The unperturbed eigenenergies (E_{v_i,J_i}) and the ac Stark shifts ($\Delta E_{v_i,J_i}$) of the initial levels as well as the photofragment recoil energy E_0' corresponding to the peak energy E_0 [Eq. (1)] are also shown.

v_i,J_i	E_{v_i,J_i} (eV)	$\Delta E_{v_i,J_i}$ (eV)	E_0' (eV)	$P_i(\theta,E_0,\Lambda,n)$ For relevant final channels (Λ,n) at $\theta=0°$ and $\theta=90°$				$\mathfrak{R}$
				$\lvert 2p\sigma_u,-1\rangle_0$	$\lvert 3d\sigma_g,-4\rangle_0$	$\lvert 2p\sigma_u,-1\rangle_{90}$	$\lvert 3d\sigma_g,-4\rangle_{90}$	$\dfrac{\sum_{\Lambda,n} P_i(\theta,E_0,\Lambda,n)_{90}}{\sum_{\Lambda,n} P_i(\theta,E_0,\Lambda,n)_0}$
4,0	−1.9566	−0.0397	0.20	2.44	2.16		0.21	0.046
4,1	−1.9535	−0.0500	0.19	1.05	6.20	0.008	0.61	0.085
5,0	−1.7921	−0.0337	0.37	0.062	4.00		0.046	0.011
5,1	−1.7891	−0.0337	0.37	1.63	8.98	0.028	0.12	0.014

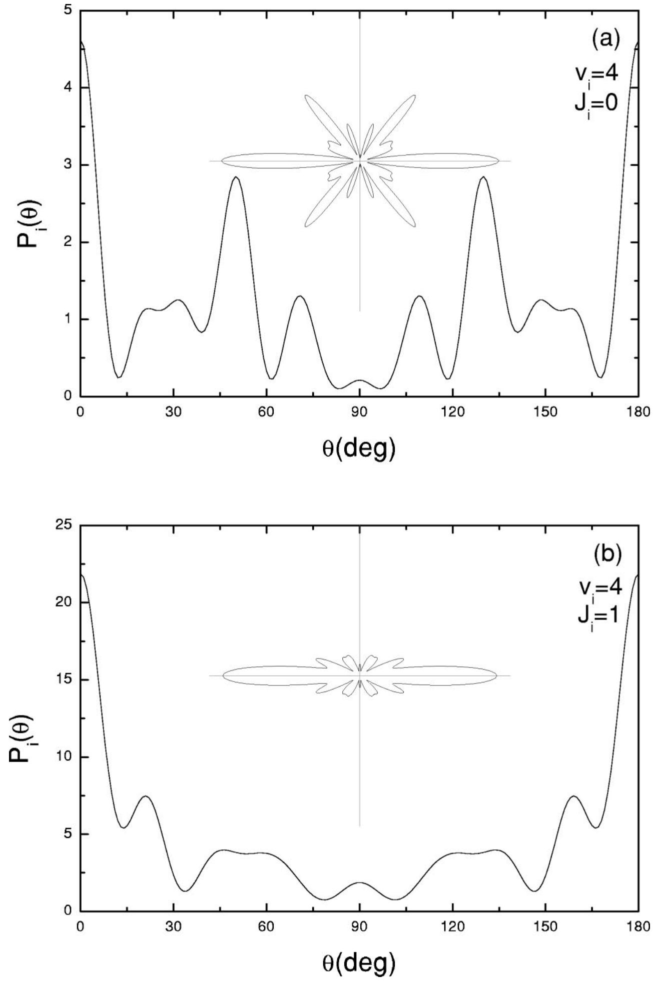

FIG. 3. Angular distributions, in both linear and polar plots, of the photofragments of D_2^+ at laser wavelength 400 nm and intensity 3×10^{13} W/cm^2 for initial levels $v_i=4$ and (a) $J_i=0$, (b) $J_i=1$.

potential energy curves followed by the system. These curves have been obtained by diagonalizing the Hamiltonian with the basis set containing all the possible rotational quantum numbers for each electronic state. However, only the states correlating to $J=0$ (for g states) and $J=1$ (for u states) have been drawn. In a more realistic representation, each of the curves should be replaced by a band of curves, which would avoid the crossings in much more involved ways. However, what is important in the present context is that each of these curves will have mixed angular momentum character.

Once a perpendicular transition occurs to the $2p\pi_u$ state, the nonadiabatic paths through the avoided crossings will be preferred because further couplings of the field with the molecule in a near perpendicular direction would be either extremely weak or nonexistent. Thus, in this case, the molecule will have the maximum probability of ending up in the state $3d\sigma_g$ with an energy corresponding to the absorption of (net) four photons. From the Table I, we find that dissociation parallel to the laser polarization from $v_i=4$, $J_i=1$ and $v_i=5$, $J_i=0$, 1 occurs predominantly in the $|3d\sigma_g,-4\rangle$ state and to a lesser extent in the $|2p\sigma_u,-1\rangle$ state. We also see that the five-photon perpendicular transition from the $|1s\sigma_g,0\rangle$ state to the $|2p\pi_u,-5\rangle$ state is not effective for the $v_i=5$ level. We also note that the branching ratios to different photon channels and the ratios $\mathfrak{R}$ of dissociation in the perpendicular direction to that in the parallel direction depend strongly on the initial vibrational-rotational levels of the molecule. Since the rotational levels and their couplings cannot be clarified on our simplified diagram (Fig. 2), the physical cause behind this variation cannot be explained with reference to this diagram.

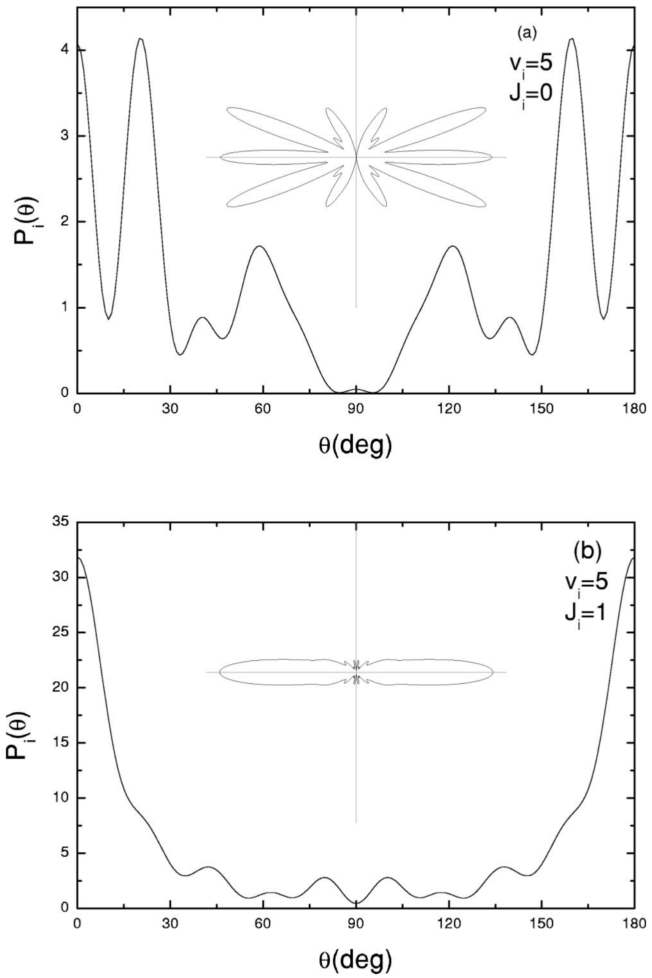

FIG. 4. Same as Fig. 3, but for $v_i=5$.

The ratios $\mathfrak{R}$ between the D^+ ions ejected in directions perpendicular and parallel to the laser polarization have also been shown in Table I for the four initial vibrational-rotational levels (v_i,J_i). This ratio is about 4.6% for $v_i=4,J_i=0$, while for $v_i=4,J_i=1$ it is about 8.5%. However, for $v_i=5$ with $J_i=0$ and 1, the ratios are about 1.1% and 1.4%, respectively. In their experiment, Chin and co-workers [7] have obtained a value of about 11% for this ratio. This value is in a reasonable agreement with our result of $\mathfrak{R}$ if we take $v_i=4,J_i=1$. For this level, the total kinetic energy would be about 0.19 eV (for net four-photon absorption).

It should be worth mentioning here that we have considered the only two high lying electronic states $2p\pi_u$ and $3d\sigma_g$ as suggested by Chin and co-workers [7] to interpret their experimental results. The consideration of the above two states made our computations feasible and at the same time made our understanding and interpretation of the dynamics of perpendicular transition through our computed results very transparent. However, the oscillator strength from the ground $1s\sigma_g$ state would certainly be distributed over other higher Σ and Π states, also. For a more accurate picture of the dynamics, other higher states should be included in the basis set. However, our computation does show convincingly that the main experimental feature obtained is indeed due to the presence of high lying electronic states.

For five-photon direct absorption from $v_i=5$ $(J_i=0)$ level, the released total kinetic energy will be about 3.2 eV as predicted by Chin and co-workers [7]. However, we have found that (net) five-photon transition does not contribute to the dissociation in the perpendicular direction. Though the five-photon coupling of the ground $1s\sigma_g$ state with the high lying $2p\pi_u$ state plays a key role, subsequent nonadiabatic transitions would force most of the perpendicularly dissociating molecules to the $|3d\sigma_g,-4\rangle$ state (Fig. 2). This is represented as

$$|1s\sigma_g,0\rangle \rightarrow |2p\pi_u,-5\rangle \rightarrow |3d\sigma_g,-4\rangle.$$

From the initial $v_i=4, J_i=0,1$ levels, the total kinetic energy release (for net four-photon absorption) will be about 0.20 eV, and from $v_i=5, J_i=0,1$ it will be about 0.37 eV (Table I).

We could not find any appreciable perpendicular dissociation from the $v_i=10$ $(J_i=0)$ level as suggested by the authors [7]. The energy release due to a (net) four-photon dissociation from the level would be, according to the authors [7], about 1.2 eV. However, as is evident from our diagrams (Figs. 1 and 2), the $v_i=10$ $(J_i=0)$ level would not be influenced by the crossing between the $|1s\sigma_g,0\rangle$ and $|2p\pi_u,-5\rangle$ states. We also did not find any $|1s\sigma_g,0\rangle \rightarrow |2p\pi_u,-3\rangle \rightarrow |3d\sigma_g,-4\rangle$ transition via three-photon radiative coupling of the $1s\sigma_g$ and $2p\pi_u$ states, as suggested by the authors [7]. This is mainly because the channel is closed at a wavelength of 400 nm or 25 000 cm^{-1} (Figs. 1 and 2).

The fact, that the $2p\pi_u$ state is responsible for dissociation in the perpendicular direction (though the photofragments mainly end up in the $3d\sigma_g$ state), has been computationally observed by excluding the $2p\pi_u$ state from our basis set. In this case, there is no fragmentation in the perpendicular direction of the laser field. Thus, the perpendicular component [16] of the transition dipole moment behaving as $\sin\theta$ (where θ is the dissociation angle with respect to the space fixed Z axis) is responsible for perpendicular ejection of the ions fragments. σ states only give rise to the parallel transitions [16] with moments behaving as $\cos\theta$ and give rise to parallel emission only.

The angular distributions P_i of the photofragments of the $\mathrm{D_2}^+$ for the two initial vibrational levels $v_i=4$ and 5 with $J_i=0,1$ are presented, in both linear and polar plots, in Figs. 3 and 4, respectively. For $v_i=4, J_i=1$, a considerable number of D^+ ions are ejected perpendicular to the laser field [Fig. 3(b)]. For $v_i=4, J_i=0$ [Fig. 3(a)], the number of ions emitted in the perpendicular direction is nearly half of that for $v_i=4, J_i=1$. For $v_i=5, J_i=0$ and 1 (Fig. 4), this number is negligible. A number of large "scattering rings" [17] at different intermediate angles are also observed. The multiphoton dissociation by a laser field in the presence of coupling to excited Π and Σ states does give rise to rings in the angular distribution of the fragments. These structures are sensitive to the initial vibrational-rotational wave function and dynamical features of the process.

IV. CONCLUSIONS

We have theoretically made an attempt to properly explore the recent experimental findings of Chin and co-workers [7] by considering high lying electronic states $2p\pi_u$ and $3d\sigma_g$, and high vibrational levels in multiphoton dissociation of $\mathrm{D_2}^+$ by intense high frequency laser fields. We have shown that the perpendicular dissociation of $\mathrm{D_2}^+$ is due to the (net) four-photon excitation of the ground $1s\sigma_g$ state to the $3d\sigma_g$ state through the intermediate five-photon coupling of the $1s\sigma_g$ and the $2p\pi_u$ states. Our computed value of the emission of D^+ ions in the perpendicular direction to that in the parallel direction of the laser field reasonably agrees with the experimental results of Chin and co-workers [7] at the same laser parameters.

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