Orientation of KRb molecules in a switched electrostatic field*

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(Received 4 August 2012; revised manuscript received 12 October 2012)

We theoretically investigate the orientation of the cold KRb molecules induced in a switched electrostatic field by numerically solving the full time-dependent Schrödinger equation. The results show that the periodic field-free molecular orientation can be realized for the KRb molecules by rapidly switching off the electrostatic field. Meanwhile, by varying the switching times of the electrostatic field, the adiabatic and nonadiabatic interactions of the molecules with the applied field can be realized. Moreover, the influences of the electrostatic field strength and the rotational temperature to the degree of the molecular orientation are studied. The investigations show that increasing the electrostatic field will increase the degree of the molecular orientation, both in the constant-field regime and in the field-free regime, while the increasing of the rotational temperature of the cold molecules will greatly decrease the degree of the molecular orientation.

Keywords: orientation, KRb molecule, electrostatic field

PACS: 37.90.+j, 33.57.+c, 42.50.Hz **DOI:** 10.1088/1674-1056/22/5/053701

1. Introduction

In recent years, more and more attention has been paid to molecular alignment and orientation[1–3] along with the development of molecular cooling. In a cold (< 20 mK) molecular system, most of the molecules populate in low rotational states. When a strong electrostatic field is applied, due to the interaction of the molecular dipole moments with the field, these low-J states can be converted from pinwheeling rotors into pendular librators. These states in the alignment and orientation where the molecular axises are confined to librate over a limited angular range along the field direction in the lab frame are called pendular states.[4,5] It has significant applications for the pendular state molecules in studying the molecular spectroscopy,[6–8] chemical reaction dynamics,[9,10] and surface science.[11] Recent investigations showed that the aligned or oriented molecular system can be regarded as an ideal quantum system when the density of the cold molecules reaches quantum gas regime.[12] They greatly enhance the importance of the realization of the alignment or orientation of the cold molecules.

Compared with the molecular alignment, it raises greater challenge to achieve the molecular orientation with a head-versus-tail order. The traditional method to realize the molecular orientation is mainly based on the electrostatic field, including the hexapole state selection[13] and the so-called brute force orientation techniques.[14,15] Only symmetric top molecules with very low density can be state-selected and oriented in the hexapole state-selection method by using an inhomogeneous electrostatic field. Comparatively, the brute force technique is applicable to all dipolar molecules. Those molecules with large permanent dipole moment and small rotational constant are favorable to obtain a high degree of the molecular orientation. Friedrich *et al.*[16] proposed a scheme to enhance the degree of the molecular orientation by using combined electrostatic and nonresonant laser fields, and the orientation was realized experimentally later by Sakai and his co-workers.[17,18] However, the electrostatic field is applied adiabatically and it can hardly realize the field-free molecular orientation so far. Thus, the subsequent applications of the molecular orientation are usually influenced by external fields.[19]

In the study of the laser-induced field-free molecular alignment and orientation, investigations showed that the realization of the field-free cases depends on the turn-off time of the laser field.[20] Meanwhile, it has been demonstrated that a slow turn-on and rapid turn-off nonresonant laser field can greatly enhance the degree of the molecular alignment, and the maximal degree is closely correlated with the turn-off time of the laser field.[21,22] Similarly, the field-free molecular orientation could probably be realized by rapidly switching the electrostatic field. The shortest switching time of the electrostatic field can be as short as 1 ns, even down to 800 ps in the laboratory[23] at present. Therefore, molecules with a relatively long rotational period may experience nonadiabatic interaction with such a switched electrostatic field, and the field-free molecular orientation would be realized.

In this paper, we propose the orientation of the KRb molecules by rapidly switching an electrostatic field. One reason why we choose the KRb molecule as the candidate is that it has relatively long rotational period, and thus one can readily

*Project supported by the National Natural Science Foundation of China (Grant No. 11034002), the State Key Development Program for Basic Research of China (Grant No. 2011921602), the Scientific and Technical Project of Nantong City, China (Grant No. BK2011052), and the Qing Lan Project.

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study the adiabatic and nonadiabatic processes in the rapid-switched electrostatic field. The other is that KRb quantum gas, below 350 nK, has been successfully produced in the laboratory by Ni *et al.*[24] Thus, the alignment and orientation of the KRb molecules have practical significance in the investigation on the new phenomena in a quantum regime. Our results show that the adiabatic and nonadiabatic behaviors of the KRb molecules can be realized by varying the switching times of the applied electrostatic field. Field-free molecular orientation can be obtained when a short switch-off time is applied, and the maximal degree of the molecular orientation is closely correlated with the switch-off time. Furthermore, the degree of the molecular orientation in the constant-field regime is influenced by the switch-on process of the electrostatic field. Finally, we calculate the dependence of the maximal degree of the molecular orientation on the strength of the electrostatic field and the molecular initial rotational temperature.

2. Theoretical model

We consider a linear molecule using the model of a rigid rotor. The time-dependent Schrödinger equation can be expressed as[25]

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t}=H(t)|\psi(t)\rangle, \tag{1}$$

$$H(t)=BJ(J+1)-\mu E(t)\cos\theta, \tag{2}$$

where B is the rotational constant of the molecule, J is the total angular momentum, μ is the permanent dipole moment of the molecule, θ is the angle between the molecular axis and the direction of the electrostatic field, and $E(t)$ is the strength of the electrostatic field at time t. Generally, the degree of the molecular orientation is defined to be the mean value of $\cos\theta$ (i.e., $\langle\cos\theta\rangle$). Considering the thermal equilibrium of the molecule system, $\langle\cos\theta\rangle$ should be averaged over the Boltzmann distribution of all the internal rotational levels, and expressed as

$$\langle\cos\theta\rangle=\sum_J W_J\sum_{M=-J}^{J}\langle\Psi_{JM}|\cos\theta|\Psi_{JM}\rangle, \tag{3}$$

where W_J is the Boltzmann weight factor in the form of

$$W_J=Q^{-1}g_J\exp[-BJ(J+1)/kT],$$

and Q is the partition function given by

$$Q=\sum_J g_J(2J+1)\exp[-BJ(J+1)/kT],$$

where g_J is the spin degeneracy factor.

The electrostatic field used in our simulation is defined (see Fig. 1) as[23]

$$E(t)=\begin{cases}E_0 f\left(5+10\dfrac{t-t_{\rm on}}{t_{\rm on}-t_{\rm i}}\right), & (t_{\rm i}\le t<t_{\rm on}),\\ E_0, & (t_{\rm on}\le t<t_{\rm off}),\\ E_0 f\left(5+10\dfrac{t-t_{\rm off}}{t_{\rm off}-t_{\rm f}}\right), & (t_{\rm off}\le t<t_{\rm f}),\\ 0, & (\text{elsewhere}),\end{cases} \tag{4}$$

where $f(t)=1/(1+e^{-2t})$. The electrostatic field is characterized by four parameters E_0, $\Delta t_{\rm on,i}=t_{\rm on}-t_i$, $\Delta t_{\rm off,on}=t_{\rm off}-t_{\rm on}$, and $\Delta t_{\rm f,off}=t_{\rm f}-t_{\rm off}$, representing the maximal strength of the electrostatic field, the switch-on time, constant-field time, and switch-off time, respectively. When $t_{\rm on}\le t<t_{\rm off}$, the strength of the electrostatic field remains E_0, and we call this regime as the constant-field regime. When the electrostatic field is completely switched off, the strength of the electrostatic field is zero, and we call this regime the field-free regime in the following description.

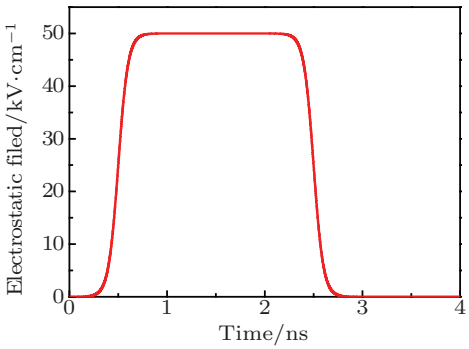


Fig. 1. (color online) Electrostatic field profile $E(t)$ with $\Delta t_{\rm on,i}=\Delta t_{\rm off,on}=\Delta t_{\rm f,off}=1$ ns, and the maximal field strength $E_0=50$ kV/cm.

In our simulation, the time-dependent Schrödinger equation is numerically solved using the split-operator method.[26] The molecular parameters of KRb are: $B=0.037$ cm^{-1}, $\mu=0.566$ D, $\alpha_{\parallel}=110.83$ Å^3, and $\alpha_{\perp}=56.68$ Å^3.[24,27] Thus, the rotational period of the KRb molecule is $T_{\rm rot}=1/(2Bc)\approx 450$ ps, where c is the velocity of the light in vacuum. The maximal strength of the electrostatic field used in the calculations is 50 kV/cm, and the initial molecular rotational temperature is 10 mK unless otherwise specified.

3. Results and discussion

It is known that the adiabatic or nonadiabatic interaction in the molecular alignment and orientation induced by the laser field depends on the ratio of the laser pulse width to the rotational period of the molecule. Similarly, the switch-on (or switch-off) time of the electrostatic field and the molecular rotational period determines the adiabatic or nonadiabatic interaction in our scheme. Considering the switching times that can be realized for dc electrostatic fields and the rotational period of the KRb molecule, we compare the molecular orientation induced by the electrostatic field with three switching

times $\Delta t_{\rm on,i} = \Delta t_{\rm f,off} = 1$, 3, and 5 ns, as shown in Fig. 2. Figure 2(a) shows the time-dependent molecular orientation for the switching time $\Delta t_{\rm on,i} = \Delta t_{\rm f,off} = 1$ ns. Here, the switching time of the electrostatic field is between $2T_{\rm rot}$ and $3T_{\rm rot}$, where $T_{\rm rot} = 450$ ps for KRb molecule. One can see that the degree of the molecular orientation is not constant and varies with the temporal evolution in the constant-field regime. This indicates that the interaction between the molecules and the electrostatic field is nonadiabatic under this switching time. In the field-free regime, the rapid switch-off process of the electrostatic field induces the generation of the rotational wave packet that periodically evolves after switching off the electrostatic field. This phenomenon is similar to the molecular alignment obtained by an ultrashort laser pulse, which is induced by the rapid interaction between the molecules and the laser field.[28] Another case of $\Delta t_{\rm on,i} = \Delta t_{\rm f,off} = 3$ ns is presented in Fig. 2(b). The interaction between the molecules and the electrostatic field tends to be adiabatic with increasing switching time. The oscillation of the degree of the molecular orientation in the constant-field regime and the maximal degree of the molecular orientation in the field-free regime are all smaller than that in Fig. 2(a). Figure 2(c) shows the case when the switching time $\Delta t_{\rm on,i} = \Delta t_{\rm f,off} = 5$ ns. Clearly, one can see that the interaction between the molecules and the electrostatic field becomes adiabatic. The degree of the molecular orientation is constant in the constant-field regime and the periodic orientation nearly vanishes after the electrostatic field is switched off. Thus, both the adiabatic orientation and the nonadiabatic orientation of the cold molecules can readily be implemented in a same experimental system by merely varying the switching time of the applied electrostatic field. Apparently, it is easier and less expensive than the multiple laser beam orientation method.

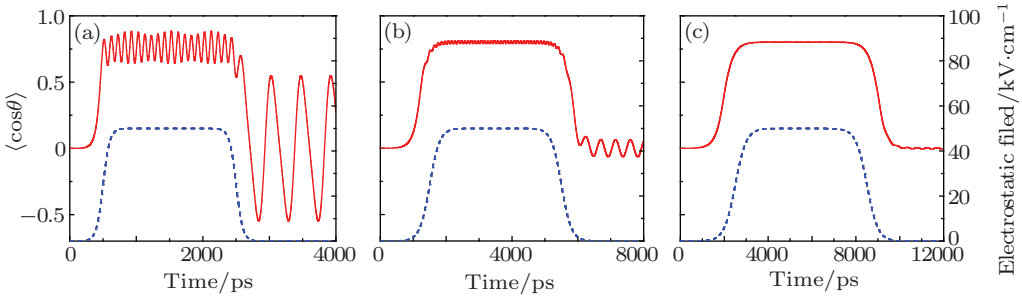


Fig. 2. (color online) Time-dependent molecular orientation $\langle \cos\theta \rangle$ (red solid lines, left axis) and the corresponding electrostatic field (blue dashed lines, right axis) for the switching times $\Delta t_{\rm on,i} = \Delta t_{\rm f,off} = 1$ ns (a), 3 ns (b), and 5 ns (c). Here, the constant-field times $\Delta t_{\rm off,on}$ are all 1 ns.

In order to analyze the degree of the molecular orientation influenced by the switch-on and the switch-off times, we investigate the temporal evolution of the molecular orientation in the field-free regime for different switch-on times at a fixed switch-off time and different switch-off times at a fixed switch-on times, and the results are shown in Figs. 3(a) and 3(b), respectively. Figure 3(a) presents the case where the switch-on times are 1, 2, 3, and 5 ns at the fixed switch-off times of 1 ns. It is shown that the field-free molecular orientation always occurs in these switch-on times, which indicates that the occurrence of the field-free molecular orientation does not evidently depend on the switch-on time of the electrostatic field. Note that the maximal degree of the molecular orientation is induced by the electrostatic field when the switch-on time $\Delta t_{\rm on,i} = 1$ ns. However, this is not similar to the results on the field-free molecular alignment induced by a slow turn-on and rapid turn-off laser field, where the slower turn-on time can increase the maximal degree of the molecular alignment.[29] The reason why the maximal degree of the molecular orientation is created with $\Delta t_{\rm on,i} = 1$ ns in our scheme is due to the nonadiabatic interaction between the molecules and the electrostatic field. In this case, the degree of the molecular orientation in the constant-field is not constant anymore and varies with time, which influences the maximal degree of the molecular orientation in the field-free regime. As the switch-on time is longer than 2 ns, the interactions between the molecules and the electrostatic field during the switch-on times become adiabatic, and the degrees of the molecular orientation in the constant-field regime nearly keep constant. Hence, the maximal degrees of the molecular orientation in the field-free regime change very little for the switch-on times of 2, 3, and 5 ns.

Figure 3(b) displays the temporal evolution of the molecular orientation in the field-free regime for different switch-off times at the fixed switch-on time of 5 ns. Now in the constant-field regime, the degree of the molecular orientation keeps constant because the switch-on process is nearly adiabatic. We can find in Fig. 3(b) that the field-free molecular orientation is closely correlated with the switch-off time. The degree of the molecular orientation decreases dramatically when the switch-off time changes from 1 to 2 ns. Furthermore, when the switch-off time of the electrostatic field is $\Delta t_{\rm f,off} = 5$ ns, the

switch-off process becomes adiabatic and the molecules go to the initial states in the absence of a field. Thus, the molecular orientation in the field-free regime almost disappears.

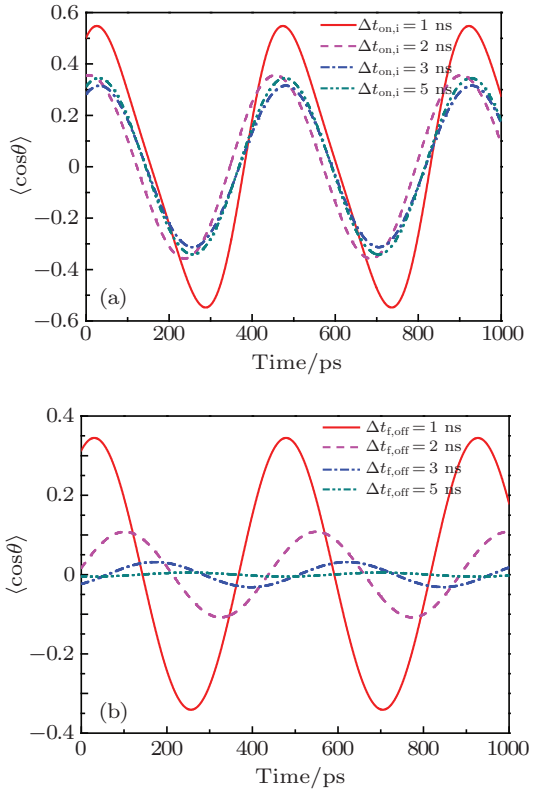


Fig. 3. (color online) Temporal evolutions of the molecular orientation $\langle\cos\theta\rangle$ in the field-free regime for (a) different switch-on times and (b) different switch-off times.

The molecular orientation influenced by the maximal strength of the electrostatic field E_0 is displayed in Figs. 4(a) and 4(b). We choose the electrostatic field with the switching times $\Delta t_{on,i} = 5$ ns and $\Delta t_{f,off} = 1$ ns, hence the field-free molecular orientation can be achieved and the degree of the molecular orientation in the constant-field regime keeps constant. Considering the experimentally available conditions, we calculate the strength of the electrostatic field up to 200 kV/cm. Figure 4(a) shows the dependence of the maximal degree of the molecular orientation on the maximal field strength in the constant-field regime. With the increase in the field strength, the maximal degree of the molecular orientation increases. This enhancement is significant at a weak applied field, and the degree of the molecular orientation gradually becomes saturated at an intense field ($E_0 > 100$ kV/cm). Here, the correlations between the strength of the electrostatic field and the molecular orientation are similar to that in the brute force technique. That is, with the increase in the strength of the electrostatic field, more rotational states are bound and the degree of the molecular orientation is enhanced.[30]

Figure 4(b) illustrates the temporal evolution of the molecular orientation for different strength of the electrostatic field in the field-free regime. Like the case in the constant-field regime, the maximal degree of the molecular orientation enhances with the increasing of the field strength. Additionally, we find that the strength of the electrostatic field can change the structure of the wave packet. Compared with that of $E_0 = 50$ kV/cm, there is obvious distinction in line configuration when $E_0 = 100$ kV/cm. This change is similar to the molecular alignment excited by a short-pulse laser field. Investigations show that the composition of the rotational wave packet is more complex with the increasing of the strength of the laser field because the number of the rotational states participating in the wave packet depends on the strength of the electric field.[31]

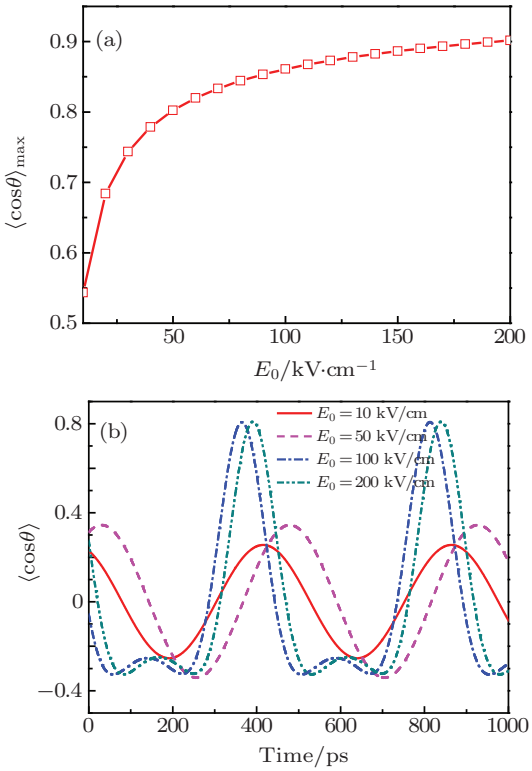


Fig. 4. (color online) (a) Maximal degrees of the molecular orientation $\langle\cos\theta\rangle_{max}$ in the constant-field regime. (b) Temporal evolutions of the molecular orientation $\langle\cos\theta\rangle$ in the field-free regime for different maximal strengths of the electrostatic field.

Additionally, we investigate the dependences of the populations of the initial rotational levels (Fig. 5(a)) and the maximal degrees of the molecular orientation in the constant-field regime (Fig. 5(b)) and in the field-free regime (Fig. 5(c)) on the temperature of the molecules. Figure 5(a) shows that molecules are populated at the ground level when the initial rotational temperature is 10 mK and the contributions to the high levels are increasing with the rising temperature. Therefore, the maximal degree of the molecular orientation in the constant-field and the field-free regimes are both decreasing with the increasing of the rotational temperature. The degree of the molecular orientation decreases sharply when the rotational temperature rises from 100 mK to 1 K, and the degree of the field-free molecular orientation with $E_0 = 50$ kV/cm turns to be zero when $T = 1$ K, as shown in Fig. 5(c). Thus,

we can conclude that low rotational temperature (less than 20 mK) is necessary for achieving a high degree of molecular orientation. Fortunately, all kinds of molecular cooling techniques (for example, photo-association of laser cooled atoms,[24] buffer gas cooling combined with low velocity filtering of polar molecules[32] and Stark deceleration of supersonic expanded polar molecules[33]) can routinely produce such cold molecules.

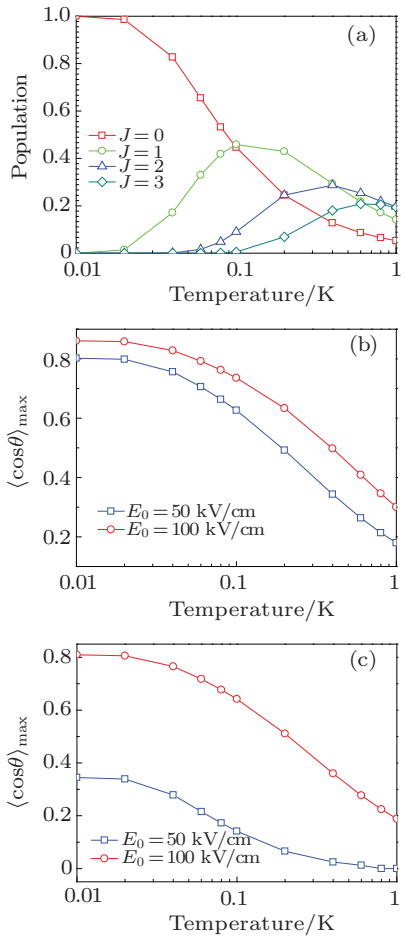


Fig. 5. (color online) (a) Population for $J \leq 3$. Maximal degrees of the molecular orientation $\langle\cos\theta\rangle_{\max}$ in (b) the constant-field regime and (c) the free-field regime as a function of the initial rotational temperature. Here, $\Delta t_{\mathrm{on,i}} = 5$ ns, $\Delta t_{\mathrm{off,on}} = \Delta t_{\mathrm{f,off}} = 1$ ns.

Oriented (especially field-free oriented) cold molecules, have many unique properties over free ones. Due to the spatial anisotropy, the oriented molecules can be used to study the anisotropic collisions of cold molecules, the external field controlled chemical reactions and thus reaction barriers, high harmonic generations, and pendular spectroscopy.

4. Conclusion

In this paper, we investigate the orientation of the cold KRb molecules in the switched electrostatic field. For such KRb molecules with a long rotational period, the simple switched electrostatic field can induce the field-free molecular orientation. The achievement of the field-free molecular orientation closely correlates with the switch-off time of the electrostatic field. The molecular orientation in the constant-field regime is also influenced by the switch-on time. If the switch-on time is much longer than the molecular rotational period, the molecular orientation in the constant-field is similar to that in the brute force technique and the interaction is adiabatic. Otherwise, a short switch-on time makes the interaction nonadiabatic and the degree of the molecular orientation vary with time. We also investigate the influences of the electrostatic field strength and the rotational temperature to the degree of the molecular orientation. The results show that both in the constant-field regime and in the field-free regime, the maximal degree of the molecular orientation can be increased with the increasing of the electrostatic field strength or the decreasing of the molecular rotational temperature.

References

[1] Stapelfeldt H and Seideman T 2003 *Rev. Mod. Phys.* **75** 543
[2] Han Y C, Hu W H, Yu J and Cong S L 2009 *Chin. Phys. B* **18** 4834
[3] Zhang S A, Lu C H, Shi J H, Jia T Q, Wang Z G and Sun Z R 2011 *Phys. Rev. A* **84** 013408
[4] Rost J M, Griffin J C, Friedrich B and Herschbach D R 1992 *Phys. Rev. Lett.* **68** 1299
[5] Kanya R and Ohshima Y 2004 *Phys. Rev. A* **70** 013403
[6] Slenczka A 1999 *Chem. Eur. J.* **5** 1136
[7] Kisiel Z, Kosarzewski J, Pietrewicz B A and Pszczółknowski L 2000 *Chem. Phys. Lett.* **325** 523
[8] Kanya R and Ohshima Y 2004 *J. Chem. Phys.* **121** 9489
[9] Wang J, Liu F, Yue D G, Zhao J, Xu Y, Meng Q T and Liu W K 2010 *Chin. Phys. B* **19** 123301
[10] Rahim M Abd El, Antoine R, Broyer M, Rayane D and Dugourd Ph 2005 *J. Phys. Chem. A* **109** 8507
[11] Jaroń-Becker A, Becker A and Faisal F H M 2003 *J. Phys. B* **36** 375
[12] Jin D S and Ye J 2011 *Phys. Today* **5** 27
[13] Parker D H and Bernstein R B 1989 *Ann. Rev. Phys. Chem.* **40** 561
[14] Friedrich B and Herschbach D R 1991 *Nature* **353** 412
[15] Franks K J, Li H Z and Kong W 1999 *J. Chem. Phys.* **110** 11779
[16] Friedrich B, Nahler N H and Buck U 2003 *J. Mod. Opt.* **50** 2677
[17] Sakai H, Minemoto S, Nanjo H, Tanji H and Suzuki T 2003 *Phys. Rev. Lett.* **90** 083001
[18] Tanji H, Minemoto S and Sakai H 2005 *Phys. Rev. A* **72** 063401
[19] Mayle M, González-Férez R and Schmelcher P 2007 *Phys. Rev. A* **75** 013421
[20] Ortigoso J, Rodríguez M, Gupta M and Friedrich B 1999 *J. Chem. Phys.* **110** 3870
[21] Sugawara Y, Goban A, Minemoto S and Sakai H 2008 *Phys. Rev. A* **77** 031403
[22] Muramatsu M, Hita M, Minemoto S and Sakai H 2009 *Phys. Rev. A* **79** 011403
[23] Sánchez-Moreno P and González-Férez R 2007 *Phys. Rev. A* **76** 053413
[24] Ni K K, Ospelkaus S, de Miranda1 M H G, Pe'er A, Neyenhuis B, Zirbel J J, Kotochigova S, Julienne P S, Jin D S and Ye J 2008 *Science* **322** 231
[25] Henriksen N E 1999 *Chem. Phys. Lett.* **312** 196
[26] Zhang S A, Shi J H, Zhang H, Jia T Q, Wang Z G and Sun Z R 2011 *Phys. Rev. A* **83** 023416
[27] Deiglmayr J, Aymar M, Wester R, Weidemüller M and Dulieu O 2008 *J. Chem. Phys.* **129** 064309
[28] Stapelfeldt H 2003 *Eur. Phys. J. D* **26** 15
[29] Yan Z C and Seideman T 1999 *J. Chem. Phys.* **111** 4113
[30] Kong W and Bulthuis J 2000 *J. Phys. Chem. A* **104** 1055
[31] Seideman T 2001 *J. Chem. Phys.* **115** 5965
[32] van Buuren L D, Sommer C, Motsch M, Pohle S, Schenk M, Bayerl J, Pinkse P W H and Rempe G 2009 *Phys. Rev. Lett.* **102** 033001
[33] Crompvoets F M H, Jongma P T, Bethlem H L, van Roij A J A and Meijer G 2002 *Phys. Rev. Lett.* **89** 093004