Combination of multiple tools for surface manipulation of polar molecules*

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A scheme of surface manipulation and control of polar molecules is proposed, which combines three tools of electrostatic velocity filtering, bunching, and storing. In the scheme, a slow molecular beam is produced from an effusive beam by surface velocity filtering. Then the velocity spread of the slow molecular beam is compressed by a buncher consisting of a series of electrodes. Following that the molecular beam with a narrow velocity spread is stored in a storage ring. Using ND_3 molecule as a tester, the feasibility of our scheme is analyzed theoretically and verified via numerical simulations that cover all three manipulation processes. The results show that cold molecular samples can be prepared from a thermal gas reservoir and stored in the storage ring with more than 10 round trips. Our combined scheme facilitates the production and manipulation of polar molecules, offering new opportunities for basic research and intriguing applications such as quantum information science and cold collisions.

Keywords: electrostatic surface manipulation, cold molecular beam

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1. Introduction

Cold molecules is a fast-growing research area motivated by many fascinating possibilities in fundamental studies and applications like cold collisions[1] and precision measurements.[2] Various approaches have been demonstrated to obtain cold molecules, including buffer gas cooling,[3] electrostatic Stark deceleration,[4,5] Zeeman deceleration,[6] laser cooling,[7,8] photoassociation,[9] and so on. Among them, electrostatic velocity filtering is a simple method that allows for selecting slowly moving molecules from a thermal gas reservoir. This idea was first proposed in the 1950s,[10] however, it was not experimentally successful until 1999. In 1999, Ghaffari *et al.* obtained a slow lithium atomic beam from a gas reservoir by using a low-pass velocity filter made of permanent magnets.[11] In 2003, Rempe's group successfully selected slow polar molecules out of a room-temperature reservoir via a bent electrostatic quadrupole.[12] In 2010, Bertsche *et al.* demonstrated a curved electrostatic hexapole[13] that can also be used to prepare slow polar molecules. The velocity-selected molecular beam has been applied to various experiments such as ion–molecule collisions[14] and cold molecule collisions.[15]

A cold molecular beam with a certain forward velocity can be confined in a storage ring, which permits the beam to repeatedly appear at certain times and positions. In 1997, Katz proposed a design of an electrostatic storage ring for polar molecules,[16] which was demonstrated by Crompvoets *et al.* in 2001.[17] Subsequently, a sectioned storage ring and a synchrotron for polar molecules were demonstrated,[18–20] which greatly increase the number of round trips of the beams. Most recently, we proposed a scheme of synchrotron allowing for controlling heavy polar molecules.[21]

In this paper, we present a combined scheme which integrates multiple manipulation tools on a substrate, including an electrostatic velocity filter, a buncher, and a storage ring. Compared to the existing manipulation tools for polar molecules, our design has the following advantages. (i) Our scheme includes both functions of production and manipulation of cold polar molecules; (ii) The scheme is based on a substrate. In other words, the cold molecules are produced and controlled in the vicinity of a substrate, which holds promising applications such as quantum computing science and the studies of molecule–surface interaction; (iii) The compact design permits them to integrate on a chip when scaling down the geometry size.

This paper is arranged as follows. Firstly, the design of multiple manipulation tools for polar molecules is introduced, and each function element is detailed one-by-one. Secondly, the feasibility of our scheme is analyzed theoretically and verified using the method of Monte–Carlo simulation with the sample molecule ND_3. The main results and conclusions are given in the end.

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2. Electrostatic surface scheme

Our design, including the electrostatic velocity filter, the buncher, and the storage ring, is shown in Fig. 1. The cross sections of the three parts are the same and illustrated in Fig. 2. A cylindrical electrode with radius r_1 is half embedded in an insulating substrate with a thickness of a, which is supported by a grounded metal plate. Over the cylindrical electrode, there is another cylindrical electrode with radius r_2. The center-to-center distance between the two cylindrical electrodes is b. The substrate is made of Al_2O_3 ceramic (relative permittivity $\varepsilon_r = 9.8$). The metal plate and the electrodes are made of stainless steel.

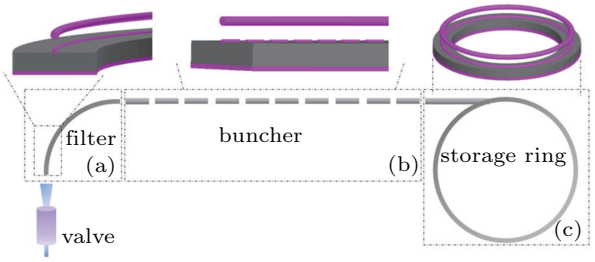


Fig. 1. (color online) Schematic of our design (top view), together with zoom-in of each element including (a) the electrostatic velocity filter, (b) the buncher, and (c) the storage ring. The molecular beam is manipulated and controlled in the vicinity of the surface of the substrate.

An effusive beam is produced by a pulse valve and then is loaded into the velocity filter which allows for preparing a slow molecular beam in the vicinity of the surface of the substrate. Subsequently, the slow molecular beam is compressed in velocity space by the buncher and finally confined in the storage ring. The heights of the potential well minima in the three manipulation tools, which can be adjusted by changing the voltages applied on them, are almost the same. Therefore, the loading efficiency of molecules among the these parts can get to a maximum. The three manipulation tools are integrated together that ensures minimizing the loading loss among them. In the following sections, the details of each element will be given one-by-one. The test parameters of the scheme are listed in Table 1.

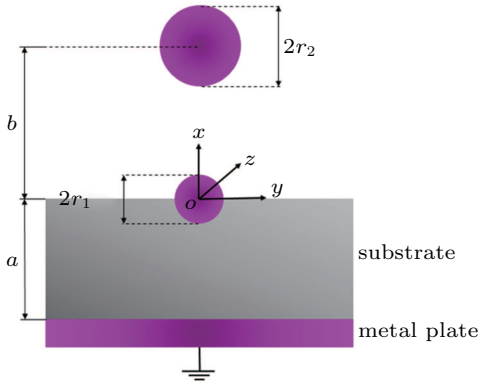


Fig. 2. (color online) Cross section of the velocity filter, the buncher (in certain positions), and the storage ring. The origin of the coordinates coincides with the center of the cylindrical electrode, where the z-axis represents the longitudinal direction of the molecular beam.

Table 1. Test parameters of the scheme.

Parameters	Symbol	Value/mm
Thickness of the substrate	a	5.0
Radius of the lower electrode	r_1	0.5
Radius of the upper electrode	r_2	1.0
Center-to-center distance between the lower and upper electrodes	b	4.5
Curvature radius of the filter	R_f	100
Length of the electrode in the buncher	l	5.0
Gap between adjacent electrodes in the buncher	d	2.0
Center-to-center distance of the buncher electrodes	L	7.0
Curvature radius of the storage ring	R_s	100

2.1. Bend filter

The electric filter is formed by bending electrodes into quarter circles with a radius of curvature R_f. When the lower and upper cylindrical electrodes are respectively charged with proper positive voltages U_1 and U_2, an electrostatic guiding center (minimum of the electric field strength) can be formed for low-field-seeking (LFS) state molecules above the surface of the substrate. The analytic expression of the electric field in our scheme is difficult to obtain, therefore we turn to numerical calculations. When voltages U_1 and U_2 are respectively set as 22 kV and 30 kV, a guiding tube for polar molecules appears, with a height of 1.2 mm from the surface of the substrate, i.e., 0.7 mm above the surface of the lower cylindrical electrode, as shown in Fig. 3(a). Figure 3(a) presents the calculated electric fields distributions in the xy plane, where the interval of the contour is 9 kV/cm. Figures 3(b) and 3(c) show the electric field distributions along the line in the x-axis and along the line across the minimum in the y direction, respectively. As shown in Fig. 3(c), the space extent of the guiding tube in the y direction is around ± 2.0 mm. Note that the height of the electric field minimum can be adjusted by changing the voltage difference between U_1 and U_2.

In the presence of the electrostatic fields, the Stark potential for polar molecules is given by[22]

$$W_{\text{stark}} = -\boldsymbol{\mu} \cdot \boldsymbol{E}, \tag{1}$$

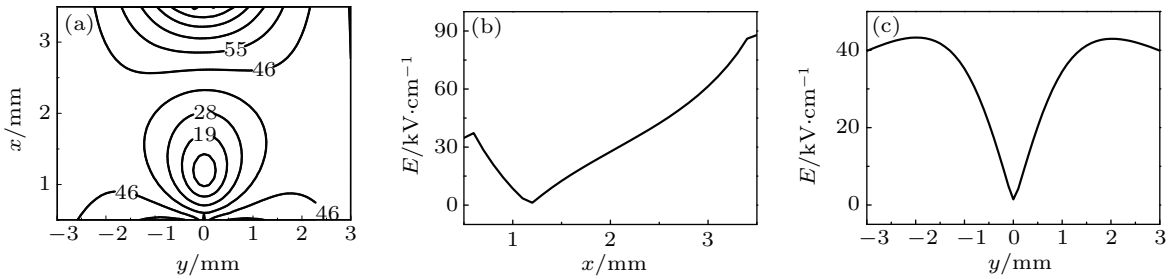


Fig. 3. (a) The contour of the electric field distributions in the xy plane under the conditions mentioned in the text. The interval of the contour is 9 kV/cm. The electric field distributions (b) along the line in the x-axis and (c) along the line across the minimum in the y direction.

where μ is the permanent electric dipole moment and E is the electric field strength. For ND_3 molecules, the Stark potential can be approximated as[23]

$$W_{\text{stark}}(r) = -\frac{MK}{J(J+1)}\mu\,|E(r)|, \tag{2}$$

where J is the total angular momentum quantum number, K and M are the projections of J on the molecular axis and the space-fixed axis, respectively. The dipole gradient force is given by

$$F = -\nabla W_{\text{stark}}. \tag{3}$$

In the bend electrostatic filter, the maximum transverse velocity is given by[24]

$$\upsilon_{\text{t,max}} = \sqrt{2W_{\text{s}}(E_{\text{max}})/m}, \tag{4}$$

where $W_{\text{s}}(E_{\text{max}})$ is the Stark shift at the maximum trapping field E_{max} and m is the molecular mass. The longitudinal cutoff velocity is given by[24]

$$\upsilon_{\text{l,max}} = \sqrt{W_{\text{s}}(E_{\text{max}})R/r_0 m}, \tag{5}$$

where r_0 is the radius of the guiding electric field. The molecules with forward velocity smaller than the maximum can be guided by the bend filter, otherwise they will overcome the trap barrier and be lost. This is exactly the principles of the bend filter for producing slow molecular beams.

2.2. Buncher and storage ring

The buncher consists of an array of cylindrical electrodes that are half-embedded in the substrate, as shown in Fig. 1(b). The length of the electrode is l, and the center-to-center distance of the electrodes is L. A gap between two adjacent segments of the cylindrical electrode has a length of d. The upper cylindrical electrode is charged with positive voltage U_3, while the odd and even electrodes half-embedded in the substrate are respectively charged with positive voltages U_4 and U_5. When these electrodes are charged with proper voltages, a train of three-dimensional electrostatic traps are formed inside the buncher. By exchanging the values of U_4 and U_5, the electric field configurations can be changed inside the buncher, as shown in Fig. 4. The solid and dashed lines in Fig. 4 indicate the two electric field distributions along z direction at a height of 1.2 mm.

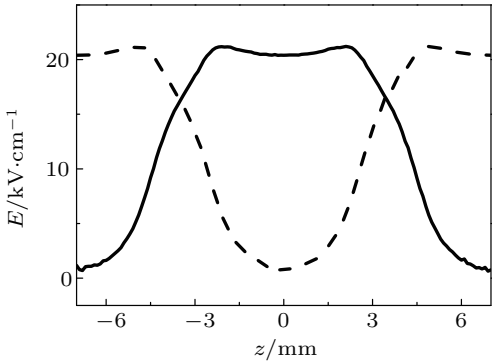


Fig. 4. Spatial distribution of the electric field (solid line) in the z direction for $x = 1.2$ mm and $y = 0$ mm. The dashed line represents the electric field when the values of U_4 and U_5 are exchanged. Geometrical parameters used for numerical calculations are shown in Table 1, and $U_3 = 10.5$ kV, $U_4 = 7.5$ kV, and $U_5 = 2.5$ kV.

By alternatively switching the two configurations in proper time, the molecular beams are bunched longitudinally. Thus the beam is kept together as a compact packet. The principle of bunching is in fact similar to that of the traditional Stark deceleration[25] with zero phase angle. During the bunching process, the molecules revolve in the phase space and get to a minimum of velocity (or space) spread at a certain time. Thus the velocity and space spread of the molecules can be controlled by adjusting the voltages applied on the electrodes and the number of the bunching stages. During the bunching process, the phase-space density remains constant according to the Liouville theorem.

Figure 1(c) displays the schematic of the storage ring. The curvature radius of the storage ring R_{s} and the voltages applied on the electrodes are the same as those of the velocity filter. Therefore, a tubular electrostatic field can be generated above the surface of the substrate inside the storage ring. The cold molecules generated from the buncher can be transversely trapped in the storage ring. The number of round trips of the beam revolving in the storage ring is proportional to $\upsilon_{\text{c}}/\Delta\upsilon$, where υ_{c} and $\Delta\upsilon$ are the longitudinal velocity and the velocity spread of the beam, respectively. The molecular packet will ultimately spread and cover the whole storage ring due to the velocity spread of the packet.

3. Monte Carlo simulations

The original molecular beam contains $\sim 4\times10^6$ ND_3 molecules in the LFS state $|J,KM\rangle = |1,-1\rangle$. The longitudinal velocity of the input molecular beam centers around $\upsilon_c = 120$ m·s^{-1} with a velocity spread (full width at half maximum, FWHM) of 130 m·s^{-1} and the transverse velocity spread is 30 m·s^{-1} in both x and y directions. These parameters are realistic from Refs. [26] and [27]. The corresponding longitudinal and transverse temperatures are respectively 7.4 K and 393 mK in the moving frame, as obtained from the formula $m(\Delta\upsilon)^2/(8\ln 2k_B)$,[28] where k_B is the Boltzmann constant. The space spreads are 20 mm and 1 mm in the longitudinal and transverse directions, respectively. Both the position and velocity spreads are Gaussian distributions. The geometrical parameters of the three elements are listed in Table 1, and the voltages used for numerical calculations are as follows: $U_1 = 22$ kV, $U_2 = 30$ kV, $U_3 = 10.5$ kV, $U_4 = 7.5$ kV, and $U_5 = 2.5$ kV. Losses due to non-adiabatic background collision are ignored in the following simulations.

The Monte Carlo simulations start from the bend velocity filter. After being filtered, the center velocity of the beam is reduced to ~ 80 m·s^{-1}, as shown in Fig. 5 (square points). At the same time, the longitudinal and transverse velocity spreads are decreased to ~ 73 m·s^{-1} and ~ 17 m·s^{-1} (not shown), respectively. The corresponding longitudinal and transverse temperatures of the resulting beam are respectively 2.3 K and 126 mK in the moving frame.

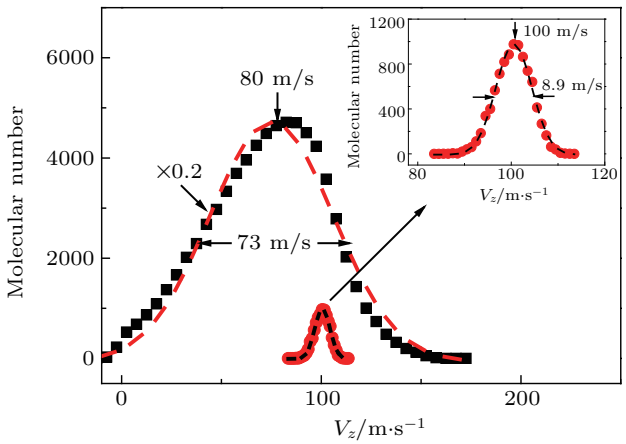


Fig. 5. (color online) Simulated longitudinal velocity distribution of the ND_3 molecular beam after the bend filter (square points). The dashed line is a fitted one and the corresponding beam has a most probable velocity of 80 m·s^{-1} with a velocity spread of 73 m·s^{-1}. Simulated longitudinal velocity distribution of the beam after 20 bunching stages (dots). It is centered around 100 m·s^{-1}, with a velocity spread of 8.9 m·s^{-1}. The dashed line is the fitting result.

Following the electric filter, the resulting beam is coupled into the buncher, where the longitudinal velocity of the synchronous molecule is set to 100 m·s^{-1}, slightly higher than the most probable velocity. After a bunching of 20 stages, the longitudinal velocity spread is greatly reduced from 73 m·s^{-1} to 8.9 m·s^{-1}, as shown in Fig. 5 (dots). The corresponding translational temperature in the moving frame is 36.2 mK.

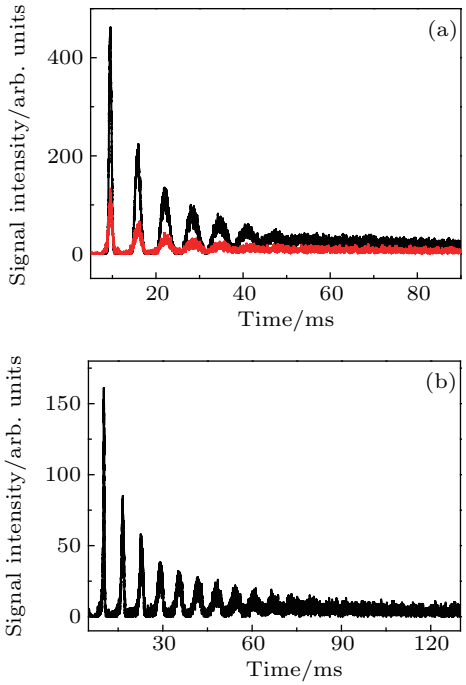


Fig. 6. (color online) (a) TOF spectra of molecular packets recorded in two storage ring schemes. The upper and lower lines represent the signal intensities of our new scheme and the previous one, respectively. (b) TOF spectra of the molecular packet under the optimal conditions. With a narrower velocity spread of the molecular packet coupled into the storage ring, the number of round trips is improved from 6 to 10.

Immediately after being bunched, the molecular packet is loaded into the storage ring and takes round trips. The TOF spectrum of the molecular packet revolved in the storage ring is indicated in Fig. 6(a) (upper line), where six round trips are recognized. The time interval between adjacent peaks is about 6.3 ms, which agrees well with the time that the molecules with a velocity of 100 m·s^{-1} take to finish one round trip in the storage ring with a curvature radius of 100 mm. By optimizing the parameters of the buncher, the velocity spread of the resulting molecular packets from the buncher can be further compressed. When $U_3 = 2.1$ kV, $U_4 = 1.5$ kV, $U_5 = 0.5$ kV, and the number of bunching stages is increased to 30, the velocity spread of the molecular packet is reduced to 5.5 m·s^{-1}, corresponding to a translational temperature of 13.2 mK. As a result, the number of round trips is increased to 10, which is shown in Fig. 6(b). In fact, our previous design of electric velocity filter presented in Ref. [29] can also be used as a storage ring. However, the optical access for that scheme is only half open, while our new design offers more optical access. In addition, our new design can offer a robust transverse force that allows for storing molecules with wider velocity spread. The trajectory calculations for both schemes are shown in Fig. 6(a). With the same initial conditions, although the numbers of round trips for the two schemes are almost the same, the number of molecules captured by our previous

scheme (lower line) is only about 40% compared to the new one (upper line).

4. Conclusions

A scheme integrating three manipulation tools, i.e., an electrostatic surface filter, a buncher, and a storage ring, is proposed and analyzed theoretically. Using the test molecule ND_3, Monte–Carlo simulations that cover all three elements are carried out to verify the possibility of our scheme. Our calculation results show that the velocity spread of an effusive beam can be greatly reduced from 130 $m{\cdot}s^{-1}$ to 5.5 $m{\cdot}s^{-1}$ after being filtered and bunched, which permits the molecular packet to make 10 round trips in our storage ring. These results show that it is effective to produce and control polar molecules in our scheme. Additionally, our scheme can offer a robust force and open optical access, which allows for preparing more cold molecules and facilitates detecting and manipulating molecules in the structure.

Our scheme can find many applications in molecular beam experiments, such as cold collisions,[1] cold chemistry,[30] high-resolution spectroscopy,[31] precision measurements,[2] and quantum optics.[32–34] The trap height above the surface of the substrate can be adjusted easily, which holds promising applications such as quantum computing science and the studies of molecule–surface interaction.

References

[1] Gilijamse J J, Hoekstra S, van de Meerakker S Y T, Groenenboom G C and Meijer G 2006 *Science* **313** 1617
[2] van Veldhoven J, Küpper J, Bethlem H L, Sartakov B, van Roij A J A and Meijer G 2004 *Eur. Phys. J. D* **31** 337
[3] Weinstein J D, deCarvalho R, Guillet T, Friedrich B and Doyle J M 1998 *Nature* **395** 148
[4] Bethlem H L, Berden G and Meijer G 1999 *Phys. Rev. Lett.* **83** 1558
[5] Liu J P, Hou S Y, Wei B and Yin J P 2015 *Acta Phys. Sin.* **64** 173701 (in Chinese)
[6] Narevicius E, Libson A, Parthey C G, Chavez I, Narevicius J, Even U and Raizen M G 2008 *Phys. Rev. Lett.* **100** 093003
[7] Shuman E S, Barry J F and DeMille D 2010 *Nature* **467** 820
[8] Zeppenfeld M, Englert B G U, Glöckner R, Prehn A, Mielenz M, Sommer C, van Buuren L D, Motsch M and Rempe G 2012 *Nature* **491** 570
[9] Miller J D, Cline R A and Heinzen D J 1993 *Phys. Rev. Lett.* **71** 2204
[10] Ramsey N 1956 *Molecular Beams* (Oxford: Oxford University Press) p. 138
[11] Ghaffari B, Gerton J M, McAlexander W I, Strecker K E, Homan D M and Hulet R G 1999 *Phys. Rev. A* **60** 3878
[12] Rangwala S A, Junglen T, Rieger T, Pinkse P W H and Rempe G 2003 *Phys. Rev. A* **67** 043406
[13] Bertsche B and Osterwalder A 2010 *Phys. Rev. A* **82** 033418
[14] Willitsch S, Bell M T, Gingell A D, Procter S R and Softley T P 2008 *Phys. Rev. Lett.* **100** 043203
[15] Sawyer B C, Stuhl B K, Yeo M, Tscherbul T V, Hummon M T, Xia Y, Klos J, Patterson D, Doyle J M and Ye J 2011 *Phys. Chem. Chem. Phys.* **13** 19059
[16] Katz D P 1997 *J. Chem. Phys.* **107** 8491
[17] Crompvoets F M H, Bethlem H L, Jongma R T and Meijer G 2001 *Nature* **411** 174
[18] Nishimura H, Lambertson G, Kalnins J G and Gould H 2004 *Eur. Phys. J. D* **31** 359
[19] Heiner C E, Carty D, Meijer G and Bethlem H L 2007 *Nat. Phys.* **3** 115
[20] van der Poel A P P, Dulitz K, Softley T P and Bethlem H L 2015 *New J. Phys.* **17** 055012
[21] Hou S Y, Wei B, Deng L Z and Yin J P 2016 *Sci. Rep.* **6** 32663
[22] Wang Q, Li S Q, Hou S Y, Xia Y, Wang H L and Yin J P 2014 *Chin. Phys. B* **23** 013701
[23] Deng L Z, Fu G B and Yin J P 2009 *Chin. Phys. B* **18** 149
[24] Motsch M, van Buuren L D, Sommer C, Zeppenfeld M, Rempe G and Pinkse P W H 2009 *Phys. Rev. A* **79** 013405
[25] Bethlem H L, Crompvoets F M H, Jongma R T, van de Meerakker S Y T and Meijer G 2002 *Phys. Rev. A* **65** 053416
[26] Tsuji H, Okuda Y, Sekiguchi T and Kanamori H 2007 *Chem. Phys. Lett.* **436** 331
[27] Hutzler N R, Lu H I and Doyle J M 2012 *Chem. Rev.* **112** 4803
[28] Gupta M and Herschbach D 1999 *J. Phys. Chem. A* **103** 10670
[29] Hou S Y, Wang Q, Deng L Z and Yin J P 2013 *J. Phys. B: At. Mol. Opt. Phys.* **46** 145303
[30] Otto R, Mikosch J, Trippel S, Weidemüller M and Wester R 2008 *Phys. Rev. Lett.* **101** 063201
[31] Hudson E R, Lewandowski H J, Sawyer B C and Ye J 2006 *Phys. Rev. Lett.* **96** 143004
[32] Ji A C, Xie X C and Liu W M 2007 *Phys. Rev. Lett.* **99** 183602
[33] Ji A C, Liu W M, Song J L and Zhou F 2008 *Phys. Rev. Lett.* **101** 010402
[34] Ji A C, Sun Q, Xie X C and Liu W M 2009 *Phys. Rev. Lett.* **102** 023602