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Orientation dependence of multiple ionization of diatomic molecules in collisions with fast highly charged ions

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We report an observation of the strong dependence of the multiple ($6<q\leqslant 10$) ionization cross section on the alignment of the molecular axis in collisions of highly charged Xe ions with N_2 molecules at an impact energy of 5.9 MeV/amu. Theoretical calculations based on the statistical-energy-deposition model well describe the experimental data.

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In collisions of fast (several mega-electron-volt/atomic mass unit) highly charged ions with molecules, multiply-ionized slow molecular fragments are copiously produced. The multiple ionization and fragmentation of simple molecules has been the subject of many recent experimental and theoretical investigations (see, e.g., [1–8] and references therein). One of the important characteristics of this process is the angular distribution of ionized fragments. In case of diatomic molecules, it is simply related to the dependence of the probability for multiple ionization on the alignment of the molecular axis with respect to the ion beam. On the basis of simple physical arguments it was expected to find highly ionized fragments preferentially produced when the molecular axis was aligned along the beam [9] so that the projectile could encounter more electrons at small impact parameters with respect to both nuclei. This idea has qualitatively been confirmed in an experimental study of 19 MeV F^{9+} collisions with N_2 molecules, producing recoil ions $N^{q_i+}, q_i\geqslant 4$ [9]. A more quantitative analysis has been performed theoretically on the basis of a simple independent atom—independent electron model [10,11]. A strong orientation effect has been predicted, with predominant multiple ionization occuring when the axis of the molecule is parallel to the projectile velocity direction. Finally, a first detailed experimental study of the orientation dependence was done for He^+ ions colliding with N_2 molecules in the energy region of 100–300 keV [12]. The predicted effect was observed for ionization multiplicity $q\geqslant 4$, and increasing with increasing q. This only involved a low-charge projectile, and it was not clear if the effect persists also for highly charged ions and larger velocities where mainly large impact parameters contribute to ionization. Some measurements for Bi-N_2 [13] and Xe-CO collisions [6] have not shown any anisotropy in the angular distributions; however, only comparatively low recoil charge states were analyzed. Moreover, calculations based on an independent electron model, with single electron-ionization probabilities calculated by the classical trajectory Monte Carlo method, have shown that for fast highly charged ions the orientation effect can hardly be larger than a few percents [6]. In order to clarify the situation, we have, therefore, carried out a series of measurements for highly charged Xe ions, accompanied by a detailed theoretical analysis.

In this paper we report the first observation of a strong orientation effect for fast highly charged ions impinging on diatomic molecules. In particular, we have studied collisions of 5.9 MeV/amu Xe^{18+} and Xe^{43+} ions with N_2 molecules. The angular distribution of $N^{q_1+}+N^{q_2+}$ recoil ions of up to $q_i=6$ was measured in coincidence. The total degree of ionization in a single event was $q=(q_1+q_2)\leqslant 10$. Details of the experimental technique have already been published [14,15]. We, therefore, give only a brief account of the experimental setup. The experiment has been performed using a 5.9 MeV/amu Xe^{18+} beam of the UNILAC accelerator at the GSI Darmstadt. An optional carbon stripper foil in front of the collision region was used to get a "Xe^{43+}" ion beam, which contained Xe^{41+} (13.5%), Xe^{42+} (18.6%), Xe^{43+} (20.2%), Xe^{44+} (16.8%), and Xe^{45+} (10.9%). The collimated beam of highly charged ions interacts with an effusive N_2-gas target. The slow ions and electrons generated in the collision process are separated by a weak homogenous electric field of 333 V/cm perpendicular to the beam and to the detector plane. Electrons are detected by a channeltron at one side of the interaction region; positive ions are accelerated towards the time- and position-sensitive multiparticle detector at the other side. After passing a field-free drift region the ions are postaccelerated to a few kilo-electron-volt to increase the detector efficiency. The detector is based on microchannel plates (MCP) in combination with an etched crossed wire anode structure consisting of independent x and y wires [14]: an electron cloud from the microchannel plates hitting a wire crossing will result in coincident pulses on the corresponding x and y wires. For each positive fragment ion the position on the detector and the time of flight relative to the electron signal are recorded. Thereby, the experimental setup is sensitive to all reaction channels resulting in at least one electron and one or more positive fragment ions. From the measured positions and flight times of the correlated fragments their initial velocity vectors can be determined using classical mechanics. If both fragments from a particular fragmen-

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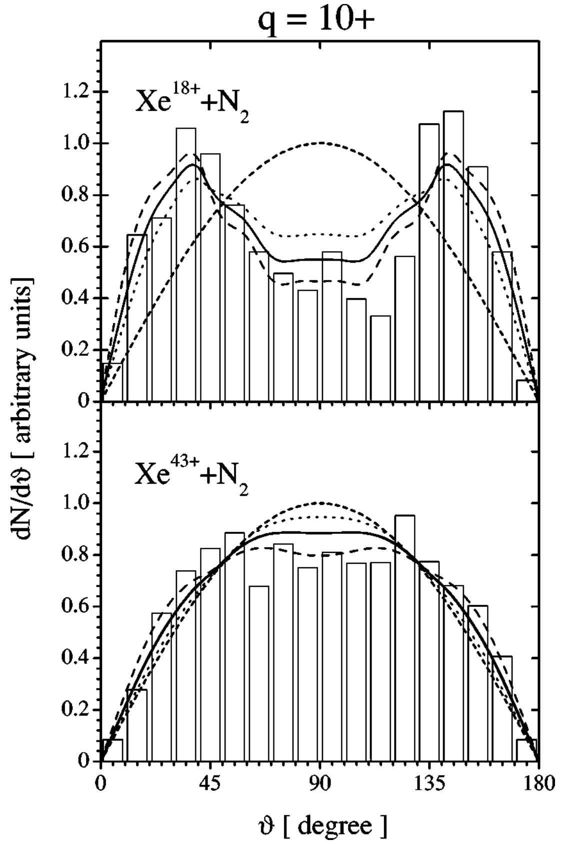

FIG. 1. Orientation dependence for the multiple ionization of N_2 molecules N_2^{10+} in collisions with 5.9 MeV/amu Xe^{18+} and Xe^{43+} ions. The histograms show the experimental results. The curves are the results of the theoretical calculations within the SED-UCA model for $g=0.005$ (solid line), $g=0.0025$ (dashed line), and $g=0.01$ (dotted line). Short-dashed curve shows the sine distribution. All curves are normalized to the same area as the experimental histograms. ϑ is the angle between the ion beam direction and the molecular axis.

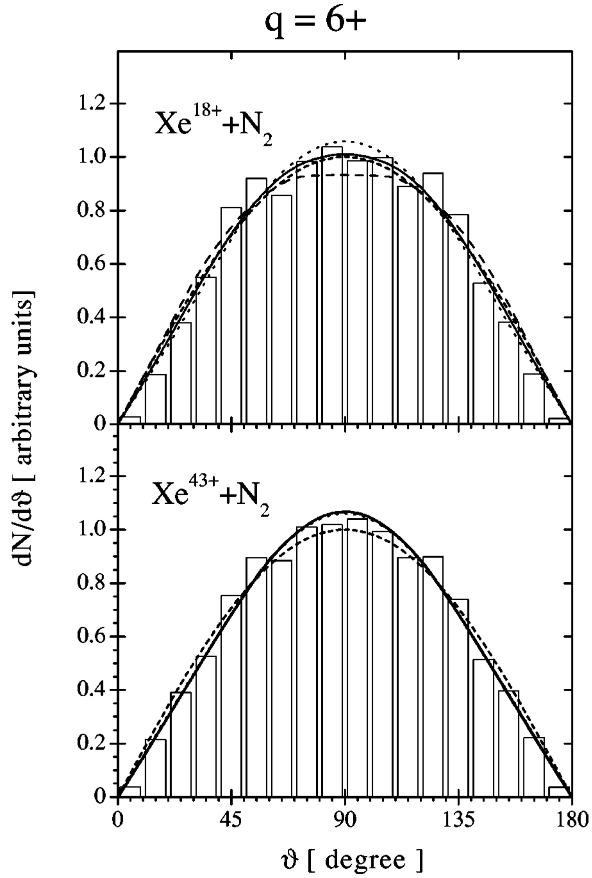

FIG. 2. The same as in Fig. 1 but for the lower degree of ionization, N_2^{6+}.

tation are detected, these data yield a kinematically complete image of the molecular breakup process, and the dissociation energy and orientation of the molecular axis can be derived for each individual event.

Examples of angular distributions are presented in Figs. 1 and 2 as histograms for the case of N_2^{10+} and N_2^{6+} ionization, respectively. The results for the N_2^{6+} ionization contains contributions from the $N^{3+}+N^{3+}$ and the $N^{2+}+N^{4+}$ channels; the N_2^{10+} ionization contains $N^{5+}+N^{5+}$ and $N^{4+}+N^{6+}$ coincidences. In both cases the symmetric fragmentation reactions mainly contribute to the distributions. The histograms in Figs. 1 and 2 should be compared with a sine function (short-dashed line) that would describe the result in case of an isotropic angular distribution. A deviation from the sine function indicates an anisotropy. Figure 1 clearly show a strong orientation effect for the Xe^{18+} projectile: the probability of tenfold ionization is suppressed when the molecule is oriented perpendicular to the beam. The effect (if any) is much smaller for the higher projectile charge Xe^{43+}. The orientation effect is practically absent for the N_2^{6+} ionization (see Fig. 2).

The revealed orientation effect and its dependence on the projectile charge and ionization multiplicity, as well as the reason of the failure of earlier attempts to find it with highly charged projectiles can be qualitatively explained as follows. An orientation effect may be expected if the effective impact parameters that mainly contribute to the ionization process, b_{eff}, is less than the dimension of the molecule: $b_{eff}\leqslant R_e$. In the opposite case, $b_{eff}\geqslant R_e$, the main contribution to the cross section comes from impact parameters much larger than the size of the molecule, and its orientation in space is not important for these distant collisions. With the increase of the projectile charge (Z_1) the strength of its interaction with electrons (characterized by the Sommerfeld parameter $\kappa=Z_1/v$) is increasing. Therefore, more electrons may be removed for a given trajectory but at the same time larger impact parameters contribute for a given degree of ionization. Thus, if a process with a small number of ionized electrons is selected in the experiment, then for highly charged ions a very wide range of impact parameters will contribute and the orientation effect will be negligible. In contrast, for a high degree of ionization the effect is observable but it should decrease with increasing projectile charge as we have observed. Based on a theoretical analysis a simple estimate is suggested in Ref. [21] for velocities $v>1$ a.u. and $\kappa>1$: the orientation effect for N_2 target can be observed if $q>5\kappa$. In our case the projectile velocity is ~ 15 a.u. Thus, for $Z_1=18$ we can expect the orientation effect for $q>6$ in accordance with our observation while for $Z_1=43$ we get $q>14$.

A quantitative theoretical interpretation of the experiment has been given within the framework of the statistical-energy-deposition (SED) model as developed by Russek-Meli [16] and Cocke [17]. We have developed an advanced version of this model [5,18] and used it for a description of the orientation dependence of the multiple ionization in He^+-N_2 collisions. Essentially, the model considers the process of multiple ionization as proceeding in two steps. First, part of the kinetic energy of the fast projectile is transferred to the target electrons. The deposited energy may be calculated using the well-developed approaches of the stopping-power theory. In the second step, when the projectile is already far away from the target, electrons are emitted, their number being proportional to the volume of the available phase space. This simple statistical approach predicts rather well the cross sections for multiple ionization in collisions of ions with atoms and molecules [5,18]. Note that a simple application of the earlier developed SED model to the case of highly charged ion collisions with molecules is not possible since a linear-response local-density approximation model [19] was used for the calculation of the deposited energy. It is based on the first-order perturbation approach that is obviously not valid for the highly charged projectile ions in the considered velocity range.

In order to overcome this difficulty we have modified the first part of the SED model calculations. We have applied the so-called unitary convolution approximation (UCA) by Schiwietz and Grande [20] to calculate the impact-parameter dependence of the electronic energy loss of fast Xe ions. Details of the application of the UCA to molecular targets are given elsewhere [21]. Here we only mention that the UCA is an approximate method that includes the Bloch higher-order corrections to the stopping power, making it applicable to highly charged ions. The further calculations within the SED model are identical to those described earlier [5]. The results of the calculations are shown in Figs. 1 and 2 as curves. Since the SED model contains one adjustable parameter, the g factor, we show results for three different values of the g factor. The solid lines show the best results, obtained for $g=0.005$. Note that this value is very close to what was used previously for the description of multiple ionization in collisions of fast ions with molecules [5]. Apparently, the SED model in combination with the UCA approach for the calculation of the energy deposition describes very well the orientation effect in Xe^{18+} and its decrease with decreasing degree of ionization and increasing projectile charge.

In summary, we have reported the first observation of the strong effect of molecular orientation in multiple ionization of diatomic molecules by fast highly charged ions. The molecules are predominantly ionized when the molecular axis is aligned along the ion beam. The effect occurs at much higher degrees of ionization then in experiments with low-charge projectiles. It decreases with increasing projectile charge and with the decrease of the degree of ionization. We have given a qualitative explanation of the observation as well as quantitative theoretical calculations; they are based on the statistical-energy-deposition model and give good description of the experiment.

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[1] U. Werner, K. Beckord, J. Becker, and H.O. Lutz, Phys. Rev. Lett. **74**, 1962 (1995).
[2] B. Siegmann, U. Werner, and H.O. Lutz, Aust. J. Phys. **52**, 545 (1999).
[3] R.L. Watson, G. Sampoll, V. Horvat, and O. Heber, Phys. Rev. A **53**, 1187 (1996).
[4] V. Krishnamurthi, I. Ben-Itzhak, and K.D. Carnes, J. Phys. B **29**, 287 (1996).
[5] N.M. Kabachnik, V.N. Kondratyev, Z. Roller-Lutz, and H.O. Lutz, Phys. Rev. A **57**, 990 (1998).
[6] L. Adoui, C. Caraby, A. Cassimi, D. Leliévre, J.P. Grandin, and A. Dubois, J. Phys. B **32**, 631 (1999).
[7] R.D. DuBois, T. Schlathölter, O. Hadjar, R. Hoekstra, R. Morgenstern, C.M. Doudna, R. Feeler, and R.E. Olson, Europhys. Lett. **49**, 41 (2000).
[8] B. Siegmann, U. Werner, R. Mann, N.M. Kabachnik, and H.O. Lutz, Phys. Rev. A **62**, 022718 (2000).
[9] S.L. Varghese, C.L. Cocke, S. Cheng, E.Y. Kamber, and V. Frohne, Nucl. Instrum. Methods Phys. Res. B **40/41**, 266 (1989).
[10] K. Wohrer and R.L. Watson, Phys. Rev. A **48**, 4784 (1993).
[11] C. Caraby, A. Cassimi, L. Adoui, and J.P. Grandin, Phys. Rev. A **55**, 2450 (1997).
[12] U. Werner, N.M. Kabachnik, V.N. Kondratyev, and H.O. Lutz, Phys. Rev. Lett. **79**, 1662 (1997).
[13] U. Brinkmann, A. Reinköster, B. Siegmann, U. Werner, H.O. Lutz, and R. Mann, Phys. Scr. **80**, 171 (1999).
[14] J. Becker, K. Beckord, U. Werner, and H.O. Lutz, Nucl. Instrum. Methods Phys. Res. A **337**, 409 (1994).
[15] U. Werner, J. Becker, T. Farr, and H.O. Lutz, Nucl. Instrum. Methods Phys. Res. B **124**, 298 (1997).
[16] A. Russek and J. Meli, Physica (Amsterdam) **46**, 222 (1970).
[17] C.L. Cocke, Phys. Rev. A **20**, 749 (1979).
[18] N.M. Kabachnik, V.N. Kondratyev, Z. Roller-Lutz, and H.O. Lutz, Phys. Rev. A **56**, 2848 (1997).
[19] J. Lindhard and M. Scharff, Mat. Fys. Medd. K. Dan. Vidensk. Selsk. **27**, 1 (1953).
[20] G. Schiwietz and P.L. Grande, Nucl. Instrum. Methods Phys. Res. B **153**, 1 (1999).
[21] Z. Kaliman, N. Orlić, N. M. Kabachnik, and H.O. Lutz Phys. Rev. A **65**, 012708 (2002).