Observation of the influence of intermolecular interactions on stopping cross sections

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The electronic stopping cross sections for $200-840$-keV ^{7}Li projectiles measured in liquid organic substances are analyzed with respect to the influence of intermolecular interaction. A systematic deviation from the average behavior has been detected. A comparison with the stopping cross sections of gases leads to the conclusion that the deviations are caused by intermolecular interactions. This is confirmed by analyzing other quantities depending upon intermolecular interactions, i.e., the van der Waals coefficients and the velocity of sound.

I. INTRODUCTION

We have measured the stopping cross sections for $200-840$-keV ^{7}Li projectiles in a large number of liquid organic compounds belonging to several hydrocarbon and alcohol series.[1] The analysis[2] of these data resulted in the decomposition of molecular stopping cross sections into partial stopping cross sections of characteristic molecular groups which are obtained as average values from the data of the associated series. It was shown that these partial stopping cross sections are additive within $1-2\%$ in analogy to a modified Bragg's rule, valid here for the characteristic groups and not for atoms (which show large deviations from additivity). For some molecules, however, deviations of $1-2\%$ from their calculated average values (outside the experimental error of less than 1%) have been observed. The purpose of this paper is to demonstrate that these deviations are due to intermolecular interactions, although their strength is much less than that of a typical chemical bond.

Since the intermolecular interaction is responsible for the condensation of a gas, a "physical-state effect" in the stopping cross section may be expected, which means that the stopping cross sections of the condensed and the gaseous phase of the same compound are different.[3] During the past few years, this problem has been investigated with growing interest.[4]

There are several direct measurements of physical-state effects. According to Akhavan-Rezayat and Palmer,[5] for example, the stopping cross sections of liquid methanol, 1-propanol, dichlormethane, chloroform, and carbon tetrachloride for $1-8$-MeV He projectiles are smaller than those

of the corresponding gases by $2-6\%$ below 2 MeV. Data by Chu et al.[6] for solid argon, oxygen, and carbon dioxide with $0.5-2.0$-MeV He projectiles are about 5% smaller below 1 MeV than the stopping cross sections of the gases obtained by the Baylor group.[7,8] The stopping cross section of ice is about 10% smaller below 1 MeV than that of water vapor, according to the measurements by Matteson et al.[9] for $0.3-2.0$-MeV He projectiles. On the other hand, there exist indirect studies of the physical-state effect. Thwaites and Watt[10] have fitted data of gaseous targets and of targets in condensed phases to universal curves separately. At about 200 keV/amu the curves for the gaseous phase lie about 15% above those for the condensed phase; at higher energies they approach each other. A similar study has been reported by Ziegler et al.[11] Generally, the stopping cross sections of gases are larger than those of the corresponding solids or liquids. This is in accordance with a calculation using modified electron distributions in the solid state which results in smaller stopping cross sections than for the free atoms.[12]

A high-precision measurement (with an accuracy of about 1%) of a physical-state effect using the same experimental techniques for the determination of the stopping cross sections for both phases has not been performed up to now. Usually, the stopping cross sections of gases are determined via the projectile's energy loss in a differentially pumped gas cell whereas those of solidified (and therefore cooled) gases are measured by backscattering. The comparison of data measured by different techniques, however, does not yield very reliable results because of possible different systematic errors. For example, the physical-state effect for argon, oxygen,

and carbon dioxide disappears when the measurements of Chu et al.[6] are compared with those of Besenbacher et al.[13] and not with those of the Baylor group.[7,8] The differentiation of energy-range curves which can be obtained for gases, solids, and liquids leads to larger errors at the low-energy end. Because of these difficulties, we use indirect methods here in the investigation of physical-state effects.

Such an analysis is described in Sec. II. The measured stopping cross sections of alkane and alcohol molecules in the liquid phase are compared with their calculated average values. The same method is applied to existing data for He projectiles in gaseous hydrocarbons and alcohols. In Sec. III the structure observed in the differences of the individual values from the average series values is compared with the similar structure for other quantities which are known to be influenced by intermolecular interactions.

II. DIFFERENTIAL ANALYSIS OF STOPPING CROSS SECTIONS

The energy loss of $200-840$-keV ^{7}Li projectiles in liquid n-alkanes (from n-pentane to n-pentadecane) and 1-alcohols (from methanol to 1-undecanol) and other organic compounds[1] has been measured via the inverted Doppler-shift attenuation method[14] with a relative accuracy of about 1%. This method is not appropriate for gaseous targets. With a cryostat the stopping cross sections of liquefied n-butane (C_4H_{10}) and propane (C_3H_8) (both gaseous at room temperature) have been determined at 200 K with the same experimental procedure as described in Ref. 1. The stopping cross section of n-pentane (C_5H_{12}, liquid at room temperature) was also measured at this low temperature to connect the new data with the existing ones. Since in our experiments the Li projectiles are created via the ^{10}B$(n,\alpha)^7$Li* reaction, each target substance needs a small amount of boron (about 1 mole % of natural boron). Trimethyl borate [B(OCH$_3$)$_3$] alone, which has been used for the measurements at room temperature, precipitates, however, at 200 K. It turned out that a 1:3 mixture of trimethyl borate and ethanol (C_2H_5OH) remains soluble in the target substances at this low temperature. The results are listed in Table I, corrected for the contribution of the boron-containing admixture. As in the previous papers,[1,2] the stopping cross sections are given at Bohr's velocity ($v_B = e^2/\hbar = 2.19 \times 10^6$ m/s), as a

TABLE I. Experimental electronic stopping cross sections of liquid propane, n-butane, and n-pentane at 200 K for ^{7}Li projectiles at Bohr's velocity in atomic units (1 a.u. $= 0.762 \times 10^{-15}$ eV cm^2). The errors represent 1.5 times the statistical standard deviation. Minimum purity of all substances: 99.5%.

Compound	Formula	Stopping cross section
Propane[a]	C_3H_8	214.8 ± 3.5
n-butane[a]	C_4H_{10}	274.3 ± 4.2
n-pentane[b]	C_5H_{12}	341.7 ± 3.5

[a]Supplier: Linde AG, Höllriegelskreuth, West Germany.
[b]Supplier: Fluka AG, Buchs, Switzerland.

linear dependence of the stopping cross sections on the velocity has been observed in the whole energy range between 200 and 840 keV.

The data of Ref. 1 are used in Ref. 2 to derive stopping cross sections of well-defined molecular groups which constitute the organic compounds. For example, the molecular groups occurring in n-alkanes and 1-alcohols are the CH$_3$ group bound to one carbon atom, the CH$_2$ group bound to two carbon atoms, and the CH$_2$OH group bound to one carbon atom. The stopping cross section of the n-alkane H$_3$C(CH$_2$)$_{v-2}$CH$_3$ is given by

$$S(C_vH_{2v+2}) = 2s(CH_3) + (v-2)s(CH_2) \qquad (1)$$

and that of the 1-alcohol H$_3$C(CH$_2$)$_{v-2}$CH$_2$OH by

$$S(C_vH_{2v+1}OH) = s(CH_3) + (v-2)s(CH_2)$$

$$+ s(CH_2OH) , \qquad (2)$$

where v is the number of carbon atoms in the molecule and s means the stopping cross section of the indicated molecular group. As discussed in Ref. 2, this decomposition ensures that the influence of the bonds between the atoms of the group and those to atoms of neighboring groups is included. A further decomposition of the CH$_2$OH group cannot be performed, because of the different type of bonds involved and the strong polarity of the C$-$O bond. As in each case the data for all members of the respective series are used for the determination of the stopping cross sections of the molecular groups, they represent average values. The usefulness of this decomposition was demonstrated by predicting stopping cross sections of more complex organic molecules which agree with the measured values within $1-2\%$. If stopping

cross sections for the alkanes and alcohols are cal-
culated in the same way and compared with the
measured values, small but distinct deviations are
observed for some molecules. These differences ΔS
for the n-alkanes and the 1-alcohols are shown in
Fig. 1. The measurement of n-pentane $(\nu=5)$ at
200 K yielded a stopping cross section 1.2% lower
than at room temperature. This does not seem un-
reasonable as the average intermolecular distances
change by an appreciable amount (about 3%) in
this temperature interval, which may affect the
stopping power. A similar temperature effect is to
be expected for n-butane and propane. Therefore,
the experimental data of n-butane and propane in
Fig. 1 have been multiplied by 1.012 before sub-
tracting the calculated average values in order to
compare them with those of the other n-alkanes
(all measured at room temperature). The abscissas
of Fig. 1 are shifted by one unit because the alkane
molecule with ν carbon atoms is isoelectronic to
the alcohol molecule with $(\nu-1)$ carbon atoms.
Data for the compounds with $\nu>9$ and $\nu>8$ have
been omitted in Fig. 1 because their absolute exper-
imental errors are too large. The parallel structure
is striking.

The projectile has the same influence on the
stopping cross section of all target substances of a
given series, because all molecules always contain
the same types of atoms. Therefore, these devia-
tions from the expected average values must stem
from properties of the target alone. There are two
possibilities to explain these deviations: First, the
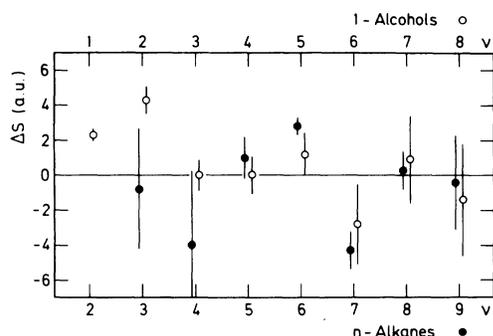structure and electron distribution of an isolated

molecule is not properly described by an average
model; although two adjacent members of each
series differ always by one CH_2 group, small irre-
gularities in the electron distribution would cause
correspondingly small differences in the stopping
power. Or, second, an isolated molecule is well ap-
proximated by the average description of its series,
but in the condensed phase a different strength of
the intermolecular interaction is responsible for the
observed behavior. The strength of the intermolec-
ular interaction is determined by the intrinsic prop-
erties of the molecules; its influence, however, is
only efficient if a considerable number of molecules
do interact, as is the case in liquids. A comparison
of data obtained in liquid and gaseous targets of
the same substances would yield quantitative infor-
mation about the influence. Unfortunately, no
stopping cross sections of gaseous alkanes (except
for methane) and alcohols for Li projectiles at
Bohr's velocity have been measured. However,
similar data exist for He projectiles. The Baylor
group has measured stopping cross sections of
gaseous methane and ethane,[8] propane and n-
butane,[15] as well as methanol, ethanol, and 1-
propanol[16] for 0.3 — 2.0-MeV He projectiles at gas
pressures of a few mbar. At 300 keV the projec-
tiles have a velocity of 3.79×10^6 m/s which is not
much larger than Bohr's velocity. No direct com-
parison is possible here, not only because of the dif-
ferent projectiles, but also because of the different
measuring methods. It is possible, however, to
analyze these data in a similar way as our Li data,
so no systematic errors are involved. To obtain
average values, a linear-regression fit according to
Eqs. (1) and (2) was performed with the alkane and
alcohol data for He projectiles and the differences
ΔS between the individual and average values are
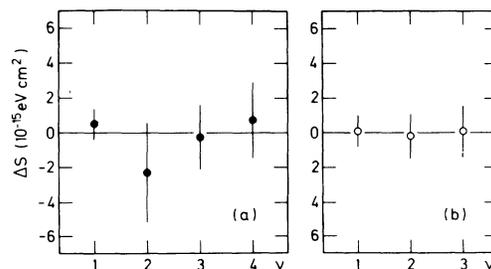plotted in Fig. 2. Evidently, a structure like that

FIG. 1. Differences ΔS between the individual experi-
mental stopping cross sections at Bohr's velocity from
Ref. 1 and Table I and the corresponding calculated
values using Eq. (1) for the n-alkanes (●) and Eq. (2) for
the 1-alcohols (○). The errors of the data (taken from
Ref. 1) represent 1 standard deviation. ν is the number
of carbon atoms, 1 a.u.$=0.762\times10^{-15}$ eV cm^2.

FIG. 2. Differences ΔS between the individual experi-
mental stopping cross sections for 300-keV He ions and
a linear-regression fit of these data; (a) for the n-alkanes
(data from Refs. 8 and 15) and (b) for the 1-alcohols
(data from Ref. 16). ν is the number of carbon atoms.

of Fig. 1 does not occur in Fig. 2. As the density in the gaseous targets is rather low, the stopping cross sections of these molecules are not influenced by a mutual interaction. Consequently, these arguments give good reasons to assume that the structure observed in Fig. 1 is caused by the interaction between neighboring molecules in the liquid.

In the condensed phase molecules attract each other via van der Waals forces (dispersion forces), as in the alkanes, for example. If the molecules have permanent dipole moments, additional electrostatic forces act; this is the case for alcohol molecules due to their OH group. Furthermore, hydrogen bonds can be formed between an OH group and an H atom of a neighboring molecule. All these interactions reduce the intermolecular distance and influence also the molecular charge distribution, which in turn may lead to an observable change in the stopping cross section. The relative contribution of hydrogen bonds is large for the small alcohol molecules methanol and ethanol, and it decreases with increasing chain length. This may be the reason for the pronounced structure in the left part of Fig. 1.

No numerical data for the magnitude of the physical-state effect and its variation exist for the series investigated here, as no stopping cross sections have been measured for Li projectiles in different phases. But no unreasonably large variations in the physical-state effect are necessary to produce a structure such as in Fig. 1. This is demonstrated by the following simple numerical example: We assume for the physical-state effect an average value of 5%, in accordance with data for other combinations of projectile and target.[5,6,9] Four hypothetical molecules of a series of organic compounds are considered, which may stand, e.g., for the cycloalkanes with five to eight CH_2 groups. Their stopping cross sections measured in the gas phase are $5s, 6s, 7s$, and $8s$, where s has a fixed value [e.g., $s = s(CH_2)$]. The contributions of the physical-state effect may oscillate around its average value by $\pm 10\%$ leading to the following stopping cross sections for the liquids:

$$S_5 = 5s(1 - 0.055),$$

$$S_6 = 6s(1 - 0.045),$$

$$S_7 = 7s(1 - 0.055),$$

$$S_8 = 8s(1 - 0.045).$$

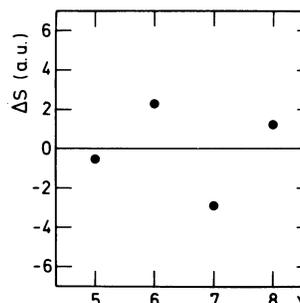The differences between these individual stopping cross sections and the corresponding average values

FIG. 3. Differences ΔS of the numerical example [calculated with $s(CH_2) = 63.4$ a.u. from Ref. 2] and a linear-regression fit of these values.

are plotted in Fig. 3. This shows that variations of $\pm 10\%$ in a physical-state effect of 5% lead to a structure similar to that found for the n-alkanes and the 1-alcohols.

So far, the above arguments support the conjecture that physical-state effects of different magnitude caused by intermolecular interactions of different strengths are responsible for the alternating structure of ΔS values in series of liquid organic compounds. In order to find further evidence for it, we have searched for physical quantities whose dependence on intermolecular interactions is well known. A comparison between the behavior of these quantities and the stopping cross sections is presented in the next section.

III. CORRELATION WITH OTHER QUANTITIES DEPENDING UPON INTERMOLECULAR INTERACTIONS

A. van der Waals coefficients

Intermolecular interactions cause deviations from the equation of state for an ideal gas. One possibility to describe real gases is given by the van der Waals equation

$$(p + a/V^2)(V - b) = N_A kT. \tag{3}$$

Here V is the volume of one mole, p the pressure, and T the absolute temperature, a and b are the van der Waals constants, N_A is the Avogadro number, and k the Boltzmann constant. The constants a and b, which are used to describe the state of the gas, will nevertheless give information about the strength of the intermolecular interaction of molecules in the liquid state. Equation (3) can be rewritten in a form resembling the virial expansion of the equation of state of real gases,

$$\frac{pV}{N_A kT} = 1 + \frac{1}{V}\left[b - \frac{a}{N_A kT}\right] + \cdots$$

The term $(b - a/N_A kT)$ is the second virial coefficient B_2, which depends in the first-order description upon the intermolecular potential U via

$$B_2 = \tfrac{1}{2}N_A \int (1 - e^{-U/kT})d^3r. \tag{4}$$

If U is approximated by a spherically symmetric Sutherland potential[17]

$$U(r) = \begin{cases} \infty, & r < r_0 \\ -Ar^{-6}, & r > r_0 \end{cases}$$

with the intermolecular distance r and constant values for A and r_0 (r_0 is here twice the molecular radius), the integration in Eq. (4) gives for $U(r) \ll kT$,

$$B_2 = \tfrac{2}{3}\pi r_0^3 N_A \left[1 - \frac{A}{kT}r_0^{-6}\right]. \tag{5}$$

Because $B_2 = b - a/N_A kT$, Eq. (5) yields

$$b = \tfrac{2}{3}\pi r_0^3 N_A \tag{6}$$

and

$$a = \tfrac{2}{3}\pi r_0^{-3} N_A^2 A \tag{7}$$

for one mole. The elimination of r_0^3 from Eqs. (6) and (7) finally gives

$$A = \tfrac{9}{4}\pi^{-2}N_A^{-3}ab . \tag{8}$$

Thus, the strength of attraction is proportional to a and b, and hence relatively large physical-state effects in the stopping power (corresponding to a reduced S) are expected for large values of a and b. If the intermolecular attraction monotonically increases with the number of carbon atoms in a series of organic compounds, plots of a and b also should be monotonic. Any oscillating structure of such plots indicates irregularities in the intermolecular potential, resulting in different physical-state effects for the individual compounds of one series.

Sufficient data for a and b exist only for the n-alkanes,[18] and are shown in Fig. 4. They obviously do not depend linearly on the number v of carbon atoms, so we used a parabolic fit to describe their average behavior (solid lines in Fig. 4). As a stronger intermolecular interaction and hence a larger product ab leads to a reduction of the stopping cross section, we plotted in Fig. 5 the differ-
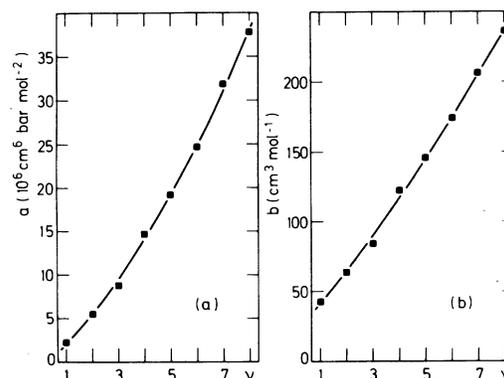

FIG. 4. The van der Waals coefficients a [Fig. 4(a)] and b [Fig. 4(b)] of Ref. 18 for the n-alkanes and their average values represented by parabolic fits (solid lines). v is the number of carbon atoms.

ences $\Delta(ab)$ between the average values and the individual values ab for the n-alkanes; the average values are again determined by a parabolic fit. Both the differences of ab and of the stopping cross sections from their average values show a remarkably parallel structure. Hence the interpretation of the structure in Fig. 1 as a result of intermolecular interactions is supported.

The stopping cross section for series of organic compounds is to first order an additive quantity, in the sense of Eqs. (1) and (2), as has been shown in Ref. 2. Therefore, it is especially useful to compare the stopping cross section with another quantity which is also additive on the average.

B. The velocity u of sound in liquids

This quantity strongly depends on intermolecular forces. Because u is not additive in a series of

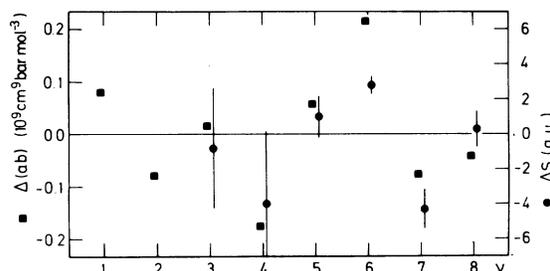

FIG. 5. Differences $\Delta(ab)$ between the average values for ab given by a parabolic fit and the individual values taken from Ref. 18 for the n-alkanes (squares, left scale). Estimated errors are 1% of ab, which is 0.002 units for $v=1$ and 0.09 units for $v=8$. For comparison, the ΔS values for the n-alkanes of Fig. 1 have been included (circles, right scale). v is the number of carbon atoms.

organic compounds, we consider instead the quantity R defined by

$$R = u^{1/3}V \tag{9}$$

which is shown in Ref. 19 to be additive. V is the mole volume of the liquid. Briefly, the explanation is based on the following arguments: In the notation used by Schaaffs,[19] u can be written as

$$u = u_\infty rs \ , \tag{10}$$

where u_∞ is an empirical constant, which has a value of 1600 m/s for many liquid organic substances including those investigated here. The velocity u depends on molecular properties and the intermolecular forces via r and s, with

$$r = B/V, \quad s = b/B \ .$$

B/N_A is the volume of one molecule, so r is that fraction of the volume V which is occupied by the molecules and s compares the van der Waals covolume b with the volume B of the molecules. For spherical, rigid molecules colliding elastically $s = 4$, but in most liquid organic compounds at room temperature $s = 2.78$.[19] Furthermore, rs always lies between 0.8 and 0.95 in liquids. Introducing $R' = Ru_\infty^{-1/3}$, Eqs. (9) and (10) give

$$R' = (rs)^{1/3}V = b^{1/3}V^{2/3} \ . \tag{11}$$

Because of $rs = b/V < 1$ and $B < b$ in liquids, the inequality

$$B < R' < V \tag{12}$$

is valid. As B and V largely behave additively, R' and therefore R are expected to be additive quantities.

It has been shown above [Eq. (8)] that the strength of the intermolecular interaction increases with increasing b. On the other hand, it decreases with increasing V. Since R' and R are dominated by the larger power of V [compared with b, Eq. (11)], R is expected to decrease when the intermolecular interaction increases. Hence, in series of organic compounds relatively large physical-state effects (corresponding to reduced stopping cross sections) should be correlated to small R values. There exist data[20] for u with an accuracy of $\leq 0.2\%$ for the series of n-alkanes, 1-alkenes, and 1-alcohols obtained with the same experimental setup. The predicted relation between R and the stopping cross section is well demonstrated in Fig. 6. For the 1-alkenes the quantity R has a similar structure, but the experimental errors of the stop-
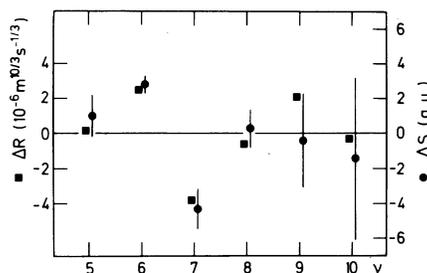

FIG. 6. Differences ΔR between the values for R representing the velocity of sound (data from Ref. 20) and a linear-regression fit of these values (squares, left scale) for the n-alkanes. The values of the densities needed for the calculation of R are taken from Ref. 21. A typical error for ΔR is one unit. For comparison, the ΔS values for the n-alkanes of Fig. 1 have been included (circles, right scale). v is the number of carbon atoms.

ping cross sections[1] are not small enough to assure the parallel behavior.

An unequivocal connection between ΔS and ΔR does not exist for the 1-alcohols. The reason for this may be that the individual alcohols of the series associate via hydrogen bonds to different degrees, besides the normal van der Waals attraction. Thus their mole volumes V clearly deviate from additivity in some cases and the same may be possible for R [Eq. (12)]. These association effects change the effective masses and, consequently, according to the molecular theory of sound,[19] the velocities of sound. But they cannot be expected to have the same influence on R and on S.

IV. CONCLUSION

It has been demonstrated that the interaction between the molecules in a liquid via dispersion and electrostatic interaction and in some cases via hydrogen bonds has an observable influence on the stopping cross section. Its absolute magnitude could not be determined, as it is at present not possible to compare values of molecular stopping cross sections measured in the gaseous and the liquid state. No measuring method with sufficient precision exists up to now, which allows the determination of molecular stopping cross sections in the gaseous, liquid, or solid state with the same procedure, as would be necessary to eliminate the influence of systematic errors.

The influence of intermolecular interactions on the stopping cross sections became apparent in clear deviations of the individual stopping cross

sections of molecules in series of liquid organic compounds from the average values determined from the data of the whole series. This observation is further confirmed by quantities whose dependence on intermolecular interaction is well known, these show the same structure when analyzed in the same way.

The differences of the stopping cross sections from their average values are of the order of $1-2$ % of the molecular stopping cross sections. No quantitative analysis of the absolute influence of the intermolecular interaction on the stopping cross section is possible from these data. However, this influence must be larger than these differences.

Consequently, more experimental data are needed to obtain quantitative results. Furthermore, it may be possible to use the measurements of stopping cross sections to investigate intermolecular interactions. The most serious implication of our results concerns the prediction of stopping cross sections; obviously, even changes of the molecular bond structure in the target due to intermolecular interactions must be taken into account.

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