Probing the helium dimer by relativistic highly-charged projectiles

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(Dated: May 7, 2021)

We study the fragmentation of He₂ dimers into He⁺ ions by relativistic highly-charged projectiles. It is demonstrated that the interaction between an ultrafast projectile with an extremely extended object – the helium dimer – possesses interesting features which are absent in collisions with “normal” molecules. It is also shown that such projectiles, due to their enormous interaction range, can accurately probe the ground state of the dimer and even be used for a precise determination of its binding energy.

1. The helium dimer, He₂, is a fascinating quantum system bound by van der Waals forces. The interaction between two ground-state helium atoms is so weak that it supports just one bound molecular state with a tiny binding energy (≈ 10⁻²⁶ eV, [1]) and an enormous size: its average bond length is ≈ 52 Å [1] while the dimer extends to the distances of more than 200 Å representing the largest known (ground-state) diatomic molecule.

Because of such extreme dimensions, the Casimir-Polder retardation effect [2] (having a relativistic origin) noticeably influences the interatomic interaction in the dimer ground state [3]. The outer classical turning point in this state is about 14 Å [3] that is almost four times smaller than its average size indicating that the dimer is a quantum halo system which spends most of the time in the classically forbidden region.

Even though the possible existence of this dimer was theoretically discussed since the 1920th, it was experimentally observed only in 1993-1994 [4-6].

2. When atomic particles (atoms, molecules) interact with each other or with external electromagnetic fields, they can be excited, ionized or even disintegrated. Ionization and fragmentation processes are of especial interest because they not only unveil valuable information about the initial system but also trigger various transformations in chemical and biological environments.

The fragmentation of the He₂ dimer into He⁺ ions induced by absorption of a single photon or by a collision with a charged projectile was explored in [7-8] and [9-10], respectively. In these papers the focus was on the fragmentation events with kinetic energies of the detected He⁺ ions ≳ 1 eV which corresponds to the start of the Coulomb explosion of the He⁺-He⁺ system at internuclear distances not exceeding ≈ 14 Å.

The authors of [11] investigated the fragmentation of He₂ into He⁺ ions either by absorption of high-frequency photons or by a strong low-frequency laser field. In both cases the dimer atoms were singly ionized within a time interval shorter than the time scale of the nuclear motion in the He-He⁺ ion, which enabled one to sample the dimer wave function and measure its binding energy.

3. Here we report on our study of the fragmentation of the helium dimer into singly charged ions by collisions with relativistic highly-charged projectiles. It will be demonstrated that the interaction of an ultrafast projectile with an extreme extended object – the helium dimer – possesses interesting (and exceptional) features which are absent in collisions with ”normal” molecules (or atoms).

It will also be shown that such projectiles, due to their ultralong interaction range, can directly probe the structure of the dimer in the halo region ≈ 14–250 Å (where it spends about 80% of the time [4]) and can be even used for an accurate determination of the binding energy of the dimer.

Atomic units (ℏ = |e| = me = 1) are used throughout unless otherwise is stated.

4. Let He₂ dimers in the ground state collide with projectiles having a charge Ze and moving with a velocity v approaching the speed of light c ≈ 137 a.u. In such collisions the parameter η = Ze/c always remains well below 1 indicating that the unitarity condition does not ”couple” different reactions which may, therefore, be considered separately. Also, the inclusion of any extra interaction step (beyond a necessary minimum) sharply reduces the production cross section. Besides, at v ≈ c electron capture processes are completely negligible compared to ionization [12]. Under such circumstances the breakup of the He₂ dimer into He⁺ ions caused by these collisions,

\[ Z_p + \text{He}_2 \rightarrow Z_p + \text{He}^+ + \text{He}^+ + e^- + e^-, \quad (1) \]

is strongly dominated by the following fragmentation mechanisms.

i. First, the projectile ”simultaneously” interacts with both atomic sites of the dimer. As a result, each helium atom emits an electron becoming a singly charged ion [13]. Since in this fragmentation mechanism the projectile directly forms the transient He⁺ - He⁺ molecular ion, while the interactions between the constituents of He₂ play here no noticeable role, we shall call it the direct fragmentation (DF). Due to the repulsion, the He⁺ - He⁺ system is unstable undergoing a Coulomb explosion.

The reflection approximation relates the kinetic energy Eker released in the explosion to the internuclear distance
$R_e$ at which it started: $E_{ker} = 1/R_e$. On the time scale of the nuclear motion in the dimer the DF mechanism leads to the sudden removal of two electrons and $R_e$ coincides with the instantaneous size $R$ of the dimer when the collision occurred.

ii. Second, the projectile interacts with just one atom of the dimer, the atom is singly ionized and the emitted electron moves towards the other atom knocking out one of its electrons. This mechanism is a combination of single ionization of a helium atom by a high-energy projectile and the so called e-2e process on helium (single ionization by electron impact) and will be abbreviated as the SI-e-2e.

In the SI-e-2e the emitted electron moves much faster than the helium nuclei. Consequently, in this mechanism (like in the DF) the energy $E_{ker}$ directly reflects the dimer size at the instant of the collision with the projectile.

iii. Third, the projectile also interacts with just one helium atom but now this results in its ionization-excitation. The residual helium ion then de-excites by transferring the energy to the other atom that leads to its ionization.

iv. The last – fourth – mechanism also involves a collision of the projectile with just one atom which, in this case, leads to its double ionization. Then the He$^{2+}$ ion radiatively captures one electron from the neutral atom.

Results on the break-up of He$_2$ dimers by photo absorption [8] and 0.15 MeV/u alpha particles [9] show that in the last two fragmentation mechanisms a very significant contraction of the He-(He$^+$)$^*$ and He-He$^{2+}$ dimers, which are directly produced in the collision, is necessary for the formation of the He$^+$-He$^+$ system. This is especially true for the mechanism, which involves the electron transfer occurring at small internuclear distances $R$ ($\lesssim 2$ Å, $E_{ker} \gtrsim 5$ eV [9]) while the range of the two-center energy exchange is mainly restricted to $R \leq 14$ Å, $E_{ker} \gtrsim 1$-2 eV [8]. In relativistic collisions, where the projectile field can be represented by "equivalent photons" [10], these mechanisms will possess same features as just mentioned above.

By comparing the fragmentation mechanisms we thus see that only in the DF and SI-e-2e the energy $E_{ker}$ is directly related to the size $R$ of the dimer at the collision instant. Therefore, unlike in the other two, in the DF and the SI-e-2e mechanisms the ground state of the dimer is directly probed. Moreover, they can be separated from the other ("interfering") two by focusing on fragmentation events with small $E_{ker}$ ($E_{ker} < 1$ eV) and in what follows we shall consider them only.

5. The DF mechanism. Its detailed theoretical description will be given elsewhere. Here we just mention that it predicts that the DF cross sections depend on the projectile charge ($\sim Z_p^4$), the impact energy (per nucleon) and the transverse size, $R_\perp = \sqrt{R^2 - (R \cdot v)^2/v^2}$, of the dimer, where $R$ is the dimer internuclear vector.

In figures [1,2] we show the cross section for the production of two singly charged helium ions by U$^{92+}$ projectiles [17]. In figure [1] the cross section is given as a function of the transverse size $R_\perp$ at different impact energies whereas in figure [2] it is plotted as a function of the impact energy for different values of $R_\perp$. Some main conclusions can be drawn from these figures.

| Collision Energy (GeV/u) | Nonrelativistic Calculation: | Relativistic Calculation: |
|-------------------------|----------------------------|--------------------------|
|                         | 1 GeV/u                    | 5 GeV/u                  |
|                         | 10 GeV/u                   | 50 GeV/u                 |

**FIG. 1**: Cross section for producing two He$^+$ ions by U$^{92+}$ projectiles via the DF as a function of $R_\perp$. (Note that at $\gtrsim 5$ GeV/u all nonrelativistic results practically coincide.)

**FIG. 2**: Same as in figure [1] but as a function of the impact energy.

First, the cross section is quite sensitive to the transverse size of the dimer varying by orders of magnitude (with the sensitivity becoming less strong when the impact energy increases). However, even for the less favourable collision geometry, when $R \perp v$ and the instantaneous size $R$ of the dimer is close to its maximal detectable value ($\approx 200 - 250$ Å, [11]), the cross section can still be of the order of 10 kb. This is surprisingly large since the projectile must irradiate an object with so enormous transverse size [18].

Second, the behaviour of the cross section on the impact energy depends on the value of $R_\perp$: at not very large $R_\perp$ the cross section (slightly) decreases with increasing the energy, whereas at very large $R_\perp$ it increases.

Third, the strength of the relativistic effects in the DF mechanism depends both on the impact energy and the value of $R_\perp$. For instance, at $R_\perp = 52$ Å the relativistic effects increase the cross section by a factor ranging from 1.42 (at 1 GeV/u) to 1.82 (at 10 GeV/u). With in-
creasing $R_{\perp}$ the relativistic effects grow and at $R_{\perp} = 100$ and 200 Å for the same impact energy range this factor varies between 2.69 and 6.24, and between 11.4 and 44.5, respectively.

At the first sight the above increases might not seem dramatic or even especially strong. They, however, are to be compared with the typical strength of relativistic effects in collisions with light atoms (or "normal" light molecules).

For instance, in the single ionization of helium atoms by high-energy projectiles the increase of the total cross section by a factor of say 1.4, 6 and 44, caused by relativistic effects, would be reached at impact energies of $\approx 14$ GeV/u, 8 x $10^{14}$ TeV/u and 1.6 x $10^{15}$ TeV/u, respectively [14]. Moreover, provided ionization is dominated by the independent interactions of the projectile with each of the "active" target electrons, the total cross section for double ionization of light atoms and molecules is essentially not influenced by relativistic effects at all, no matter how high is the impact energy [20].

We have found that at $R_{\perp} \gg 1$ the total cross section for the production of two He$^{+}$ ions via the DF can quite well be approximated by

$$\sigma^{DF} \approx C \frac{Z_p^2}{v^2 \gamma^2} \left[ K_1 \left( \frac{\pi R_{\perp}}{\gamma v} \right) + \frac{1}{\gamma^2} K_0^2 \left( \frac{\pi R_{\perp}}{\gamma v} \right) \right], \quad (2)$$

where $K_0$ and $K_1$ are the modified Bessel function [21], $\varpi \approx 1.2$ a.u. is the mean transition frequency for single ionization of a helium atom, $Z_p$ and $v$ are given in a.u. and $\sigma$ in kb and $C$ is a parameter weakly dependent on $R_{\perp}$ [22].

Eq. (2) captures all essential features of our numerical results. In particular, since at $x < 1$ $K_0(x) \sim \ln(1.12/x)$ and $K_1(x) \sim 1/x$ while at $x > 1$ $K_0(x) \sim K_1(x) \sim \sqrt{\frac{\pi}{x}} \exp(-x)$, it is easy to see from Eq. (2) that the projectile is able to efficiently irradiate both atoms of the dimer provided its transverse size $R_{\perp}$ is smaller than the adiabatic collision radius $R_a = \frac{\varpi}{\gamma v}$. For an illustration, in figure 3 $R_a$ is shown for several impact energies and compared with the extension of the dimer ground state.

Besides being strongly influenced by relativistic effects, the process of the DF of He$_2$ has yet another feature. As is known, photo ionization by a weak electromagnetic field is a purely quantum process while ionization of atoms and "normal" molecules by fast charged particles can be rather well treated by classical physics even in the weak perturbation limit.

However, when $R_{\perp} \geq 1$ a.u. a classical description of the DF completely fails underestimating the cross sections by orders of magnitude. The reason is that very distant inelastic collisions are poorly described by a classical treatment whereas the simultaneous ionization of both atoms of the dimer at $R_{\perp} \gg 1$ implies that the projectile has a very large impact parameter with respect to at least one of them.

In figure 4 we display the weighted probability, $bP(b)$, for the DF mechanism as a function of the impact parameter $b$ (counted from one of the dimer nuclei). It is seen that this probability has a pronounced two-peak structure showing that the majority of the fragmentation events occurs when the projectile passes close to either one or the other helium atom.

![FIG. 4: The weighted probability $bP(b)$ for the DF given as function of the impact parameter $b$.](image)

6. The SI–e-2e mechanism. It will be shown elsewhere that in this mechanism the cross section for the production of two He$^{+}$ ions is given by

$$\sigma^{SI–e-2e} = \frac{3 \sin^2 \vartheta_R}{4\pi R^2} \int_{E_{He}}^{\infty} \frac{da^{SI}}{dx_k} \sigma^{e-2e}(\varepsilon_k), \quad (3)$$

where $\frac{da^{SI}}{dx_k}$ is the cross section for single ionization of a helium atom by a high-energy projectile differential in the energy $\varepsilon_k$ of the emitted electron, $\sigma^{e-2e}(\varepsilon_k)$ is the cross section for the total single ionization of a helium atom by an electron incident on the atom with an energy $\varepsilon_k$, $I_{He} \approx 24.6$ eV is the helium ionization potential and $\vartheta_R = \arccos(R \cdot v/R v)$.

Eq. (3) shows that the SI–e-2e mechanism is long ranged, depending on $R$ as $R^{-2}$, but becomes inefficient when the angle between the dimer orientation and the collision velocity is small. The dependence of the cross section [3] on the projectile charge and impact energy is similar to that for single ionization of a helium atom; in particular, it is proportional to $Z_p^2$ and weakly (logarithmically) influenced by relativistic effects.

![FIG. 3: The dimer ground state, and the adiabatic radius $R_a$ at different impact energies.](image)
Our analysis shows that in collisions with ultrafast ions having very high charges the SI–e–2e is much less efficient than the DF. However, if $Z_p/e \ll 1$, the SI–e–2e becomes dominant provided the dimer orientation angle $\theta_R$ is not too small.

7. In the DF and SI–e–2e mechanisms the Coulomb explosion in the He$^+$-He$^+$ system begins when the positions of the dimer nuclei are the same as right before the collision. Hence, by measuring the $E_{\text{ker}}$ spectra produced via these two mechanisms, one could directly probe the ground state of the dimer making its instantaneous “snapshots”.

However, in the other two fragmentation mechanisms the kinetic energy release is not directly related to the dimer size at the collision instant. Therefore, in order to exclude their interference, they have to be "turned off". Being characterized by relatively large values of $E_{\text{ker}} (E_{\text{ker}} \gtrsim 1 \text{ eV})$, they can be "turned off" by simply focusing on fragmentation events with $E_{\text{ker}}$ below 1 eV.

In addition to the Coulomb explosion, the He$^+$ ions have kinetic energy from the nuclear motion before the collision: it is, however, negligible because the depth of the potential well in He$_2$ is just 1 meV. Besides, the He$^+$ ions also acquire a kinetic energy directly in the ionization process. In high-energy collisions the momentum transfer $p_{\text{He}^+}$ to the He$^+$ ions in an overwhelming majority of ionizing events does not exceed 1 a.u. with the corresponding recoil energy of $2 \times (p_{\text{He}^+}^2/2M_{\text{He}^+}) \lesssim 4$ meV. Therefore, in order that the reflection approximation $E_{\text{ker}} = 1/R$ may still be used, one must have $E_{\text{ker}} \gg 4$ meV (that corresponds to $R \ll 7 \times 10^3$ a.u.).

Thus, the DF and SI–e–2e can be used for a direct probing of the dimer ground state at 14 Å $< R \lesssim 250$ Å (corresponding to 60 meV $\lesssim E_{\text{ker}} < 1$ eV), i.e. of its halo region where the dimer spends most of the time. Using the cross section for the production of two helium tiles, for different ranges of the dimer orientation. Because of the extremely long interaction range inherent to the ultrafast projectile, the spectrum intensities remain significant even at energies corresponding to very large internuclear distances $R$.

At $R \gg 1$ the probability density of the dimer is $|\Psi|^2 \sim \exp(-2k_0 R)/R^2$, where $k_0 = \sqrt{M_{\text{He}} I_b/\hbar^2}$ with $M_{\text{He}}$ being the mass of the helium atom. A small variation $\Delta I_b$ of the dimer binding energy $I_b$ ($\Delta I_b/I_b \ll 1$) changes $|\Psi|^2$ by the factor $\exp(-k_0 R(\Delta I_b/I_b))$. This in turn affects the shape of the kinetic energy release spectrum. Since this spectrum effectively spans a very broad range of $R$, it becomes sensitive even to a very small change in the dimer binding energy.

In particular, the reported values for the binding energy vary between 44.8 neV [23] and 161.7 neV [23], with 139.2 neV [23] (used in our calculations) and 151.9 neV [11] being regarded as most precise having the relative difference of just 9%. However, in the ranges 0.1 eV $\leq E_{\text{ker}} \leq 0.8$ eV, 0.075 eV $\leq E_{\text{ker}} \leq 0.8$ eV and 0.06 eV $\leq E_{\text{ker}} \leq 0.8$ eV (the latter two are not shown in fig. 3) this 9% are already converted into, respectively, 14%, 20% and 26% difference in the shape of the energy spectrum. This suggests that collisions with ultrafast projectiles can be used for an accurate determination of the He$_2$ binding energy.

The cross section for the DF, integrated over the kinetic energy release between 0.1 and 1 eV by U$^{92+}$ projectile having impact energy $\sim 1-5$ GeV/u is of the order of $10^{-19} - 10^{-18}$ cm$^2$. This value could be compared with the total cross section for single ionization of helium atom, which is $\sim 10^{-15}$ cm$^2$, the cross section for single electron capture from helium atom, which is $\sim 10^{-23}$ cm$^2$ and the cross section for the reaction $Z_p + \text{He}_2 \rightarrow Z_p + \text{He}(1s^2) + \text{He}(1s^2)$, which (according to rough estimates) is $\sim 10^{-16}$ cm$^2$.

8. In conclusion, we have studied the fragmentation of the helium dimer into singly charged helium ions by relativistic highly-charged projectiles. It was found that the breakup events with kinetic energy release $< 1$ eV are in essence solely caused by the direct fragmentation mechanism in which the projectile "simultaneously" ionized both dimer’s atoms. It was shown that this mechanism is exceptionally strongly influenced by relativistic effects and that a classical description of the collision dynamics in this case completely fails.

It was also demonstrated that ultrafast projectiles, due to their extremely long interaction range, can directly and accurately probe the structure of the dimer ground state in the halo region $\sim (14 - 250)$ Å where the dimer spends four-fifths of the time. Moreover, a rather high sensitivity of the $E_{\text{ker}}$-spectrum to the value of the dimer binding energy suggests that such projectiles can be used for its more precise determination.

Collisions with ultrafast projectiles can also be applied to explore the ground states of $^6\text{LiHe}$ and $^7\text{LiHe}$ dimers, which are other humungous diatomic molecules having the average size of about 49 and 28 Å, respectively, [27].
Finally we note that experimental facilities, where very large dimers can be explored by collisions with ultrafast highly charged projectiles, are available. 

Acknowledgements. The authors gratefully acknowledge the support from the China Scholarship Council and CAS President’s Fellowship Initiative.

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