Probing the Helium Dimer by Relativistic Highly Charged Projectiles

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We study the fragmentation of He_2 dimers into He^+ ions by relativistic highly charged projectiles. We demonstrate that the interaction between an ultrafast projectile with an extremely extended object—the helium dimer—possesses interesting features that are absent in collisions with "normal" molecules. We also show that such projectiles, due to their enormous interaction range, can accurately probe the ground state of the dimer and even be used for a determination of its binding energy.

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The helium dimer He_2 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 ($\simeq 10^{-7}$ 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] noticeably influences the interatomic interaction in the dimer ground state [3]. The outer classical turning point in this state is about 14 Å [4], which is almost 4 times smaller than its average size, indicating that the dimer is a quantum halo system that spends most of the time in the classically forbidden region. Even though the possible existence of this dimer has been theoretically discussed since the 1920s, it was only experimentally observed in 1993–1994 [5,6].

When atomic particles 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, since they not only unveil valuable information about the initial system, but also trigger various transformations in chemical and biological environments.

The fragmentation of the He_2 dimer into He^+ ions induced by absorption of a single photon or by a collision with an ion was explored in [7,8] and [9,10], respectively, where the focus was on the fragmentation events with kinetic energies of the He^+ ions $\gtrsim 1$ eV corresponding to the start of the Coulomb explosion of the He^+-He^+ system at internuclear distances $\lesssim 14$ Å. In [11] the He_2 fragmentation into He^+ ions by absorption of two high-frequency photons was exploited to measure the dimer binding energy.

In this Letter, we consider 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 that 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 $\sim$14–250 Å (where it spends about 80% of the time [4]) and can be used for an accurate determination of its binding energy. Atomic units ($\hbar = |e| = m_e = 1$) are used throughout unless otherwise is stated.

Let He_2 dimers in the ground state collide with projectiles having a charge Z_p and moving with a velocity $\boldsymbol{v}$ approaching the speed of light $c \approx 137$ a.u. In such collisions the parameter $\eta = Z_p/v$ 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. In addition, at $v \sim c$ electron capture is negligible compared to ionization [12–14]. Under such circumstances, the He_2 breakup into He^+ ions caused by these collisions is strongly dominated by the following fragmentation mechanisms.

(i) First, the projectile "simultaneously" interacts with both atoms of the dimer. As a result, each atom emits an electron becoming a singly charged ion [15]. Since here the projectile directly forms the transient He^+-He^+ system, while the interactions between the constituents of He_2 play no noticeable role, we shall call it "the direct fragmentation" (DF). Because of the repulsion, the He^+-He^+ system is unstable undergoing a Coulomb explosion.

The reflection approximation relates the kinetic energy E_{ker} released in the explosion to the internuclear distance R_{ce} at which it started: $E_{\mathrm{ker}} = 1/R_{ce}$ [16]. On the timescale of the nuclear motion in the dimer, the DF mechanism leads to sudden removal of two electrons and R_{ce} coincides with the dimer size R at the collision instant.

(ii) Second, the projectile interacts with just one atom of the dimer, the atom is singly ionized, and the emitted electron moves toward the other atom, knocking out one of its electrons. This mechanism is a combination of single ionization of He by a projectile and the e-2e process on He (single ionization by electron impact) and will be abbreviated as SI − e-2e [17,18].

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 $E_{\rm ker}$ directly reflects the dimer size at the instant of the collision.

(iii) Third, the projectile also interacts with just one atom but now this results in its ionization excitation. The residual He^+ deexcites by transferring the energy to the other atom via interatomic Coulombic decay [19] that leads to its ionization.

(iv) The fourth—and last—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+} captures one electron from He via radiative charge transfer [20].

Results on the He_2 break up by photo absorption [8] and 0.15 MeV/u alpha particles [9] show that, in the last two fragmentation mechanisms, a significant contraction of the transitory He-$(He^+)^*$ and He-He^{2+} dimers is necessary to form the He^+-He^+ system. This is especially true for the mechanism, which involves radiative charge transfer occurring at small internuclear distances ($R \lesssim 2$ Å, $E_{\rm ker} \gtrsim$ 5–7 eV [9]), while the range of interatomic Coulombic decay is mainly restricted to $R \lesssim 14$ Å, $E_{\rm ker} \gtrsim 1$ eV [8]. All this will be the case also in relativistic collisions, where the projectile field can be represented by "equivalent photons" [21].

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

The theoretical description of the DF mechanism is presented in the Supplemental Material [22]. In particular, in the impact parameter space, the transition amplitude is given by the product of the single-atom transition amplitudes, which are calculated using the relativistic symmetric eikonal approximation (see Supplemental Material [22] for details). The theory predicts that the DF cross sections depend on the projectile charge ($\sim Z_p^4$), the impact energy (per nucleon), and the transverse size, $R_\perp = \sqrt{R^2 - (\boldsymbol{R}\cdot\boldsymbol{v})^2/v^2}$, of the dimer, where $\boldsymbol{R}$ is the dimer internuclear vector.

In Figs. 1 and 2, we show the cross section for the production of two singly charged helium ions by U^{92+} projectiles [28]. In Fig. 1, the cross section is given as a function of the transverse size $R_\perp$ at different impact energies, whereas in Fig. 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.

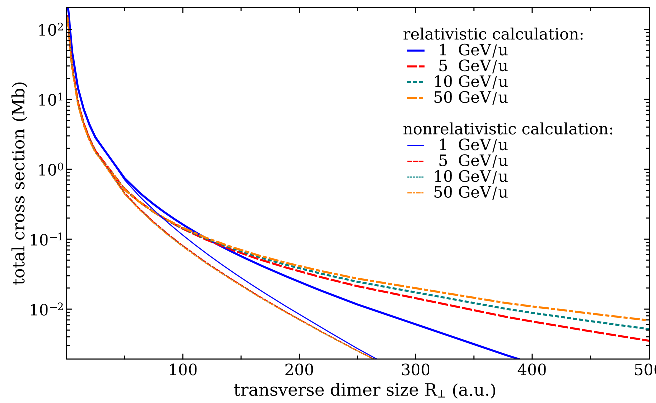

FIG. 1. Cross section for producing two He^+ ions by U^{92+} projectiles via the DF as a function of $R_\perp$. (Note that at $\gtrsim$5 GeV/u all nonrelativistic results practically coincide.)

First, the cross section is very 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 favorable collision geometry, when $\boldsymbol{R}\perp\boldsymbol{v}$ and the instantaneous size R of the dimer is close to its maximal detectable value (≃470 a.u. [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 [29].

Second, the behavior 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 $R_\perp = 100$ a.u. these effects increase the cross section by a factor ranging from 1.42 (at 1 GeV/u) to 1.82 (at 10 GeV/u). With increasing $R_\perp$, they grow and at $R_\perp = 200$ and 400 a.u., for the same

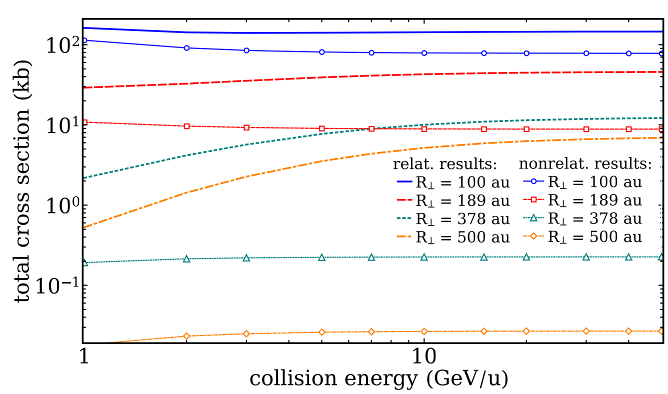

FIG. 2. Same as in Fig. 1, but as a function of the impact energy.

impact energy range, this factor varies between ≈2.7 and ≈6.3 and between ≈11 and ≈45, respectively.

At first sight, the above increases might not seem dramatic or even especially strong. However, they are to be compared with the typical strength of relativistic effects in collisions with light atoms (or normal light molecules). For instance, in 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 ≈14 GeV/u, 8×10^{11} TeV/u, and 1.6×10^{105} TeV/u, respectively [30]. 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 the impact energy [23].

At $R_\perp \gg 1$, the total cross section for the production of two He^+ ions via the DF can be approximated by [22]

$$\sigma^{\mathrm{DF}} \approx C \frac{Z_p^4}{v^4\gamma^2}\left[K_1^2\left(\frac{\bar{\omega}R_\perp}{\gamma v}\right) + \frac{1}{\gamma^2}K_0^2\left(\frac{\bar{\omega}R_\perp}{\gamma v}\right)\right], \qquad (1)$$

where K_0 and K_1 are the modified Bessel function [24], $\bar{\omega} \approx 1.2$ a.u. is the mean transition frequency for single ionization of a helium atom, and C is a parameter weakly dependent on $R_\perp$ [31].

Equation (1) captures all essential features of our numerical results. In particular, since $K_0(x) \sim \ln(1.12/x)$ and $K_1(x) \sim 1/x$ if $x < 1$, while $K_0(x) \sim K_1(x) \sim \sqrt{(\pi/2x)}\exp(-x)$ at $x > 1$, it follows from Eq. (1) that the projectile is able to efficiently irradiate both atoms of the dimer provided $R_\perp$ is smaller than the adiabatic collision radius $R_a = (\gamma v/\bar{\omega})$. As $R_a \sim \gamma v$, an ultrafast projectile possesses a very large effective interaction range that makes it an attractive tool to probe systems with large dimensions. In Fig. 3, R_a is shown for several impact energies and its comparison with the extension of the dimer ground state indicates that, beginning with impact energies of a few GeV/u, the projectile efficiently interacts with both atoms of the dimer even when it is perpendicular to the collision velocity.

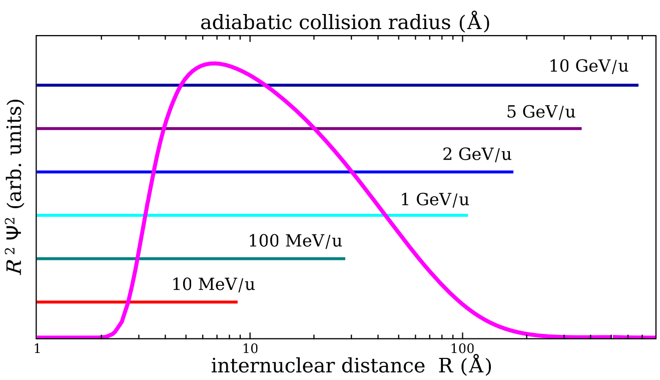

FIG. 3. The dimer ground state and the adiabatic radius R_a at different impact energies.

Besides being strongly influenced by relativistic effects, the process of the DF of He_2 has yet another feature. As is known, photoionization by a weak electromagnetic field is a purely quantum process, while ionization of atoms and normal molecules by fast charged particles can be treated with acceptable accuracy by classical physics, even in the weak perturbation limit [32].

However, when $R_\perp \gg 1$ 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 Fig. 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 sharply peaks at $b \simeq 1$ and $b \approx R_\perp$, showing that the fragmentation occurs mainly when the projectile passes close to one of the atoms. Indeed, in such a case, $bP(b)$ involves just one small factor: the probability to ionize the distant atom (that at $1 \ll R_\perp \lesssim R_a$ scales as $1/R_\perp^2$). In contrast, when the projectile has large impact parameters with respect to both atoms, $bP(b)$ involves a product of two small factors, since now both atomic ionization probabilities are small.

As it can be shown [22], in the $\mathrm{SI} - e\text{-}2e$ mechanism, the cross section for the production of two He^+ ions is given by

$$\sigma^{\mathrm{SI}-e\text{-}2e} = \frac{3\sin^2\theta_{\boldsymbol{R}}}{4\pi R^2}\int_{I_{\mathrm{He}}}^{\infty} d\varepsilon_k \frac{d\sigma^{\mathrm{SI}}}{d\varepsilon_k}\sigma^{e\text{-}2e}(\varepsilon_k), \qquad (2)$$

where $d\sigma^{\mathrm{SI}}/d\varepsilon_k$ is the cross section for single ionization of a helium atom by a high-energy projectile differential in the energy ε_k of the emitted electron, $\sigma^{e\text{-}2e}(\varepsilon_k)$ is the total

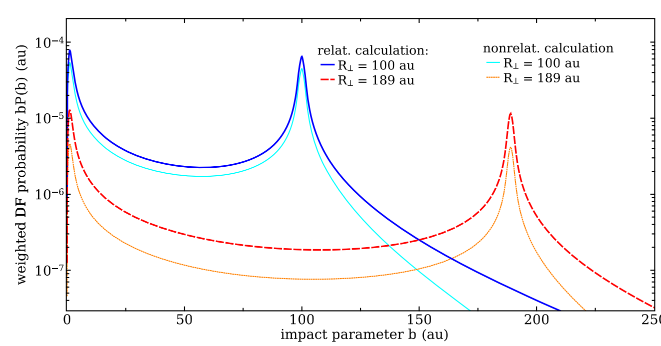

FIG. 4. The weighted probability $bP(b)$ for the DF given as a function of the impact parameter b.

cross section for single ionization of a helium atom by an electron incident with energy ε_k, $I_{\rm He} \approx 24.6$ eV is the helium ionization potential and $\theta_{\boldsymbol{R}} = \arccos(\boldsymbol{R}\cdot\boldsymbol{v}/Rv)$.

Equation (2) 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 (2) on the projectile charge and impact energy is similar to that for single ionization of a helium atom: it is proportional to Z_p^2 and weakly (logarithmically) influenced by relativistic effects.

Our analysis shows that in collisions at $Z_p/v \lesssim 1$ the SI $-$ e-$2e$ is much less efficient than the DF. However, if $Z_p/v \ll 1$, the SI $-$ e-$2e$ becomes dominant provided the dimer orientation angle $\theta_{\boldsymbol{R}}$ is not too small.

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 $E_{\rm ker}$ spectra produced via these two mechanisms, one could directly probe the dimer ground state 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, we shall focus on fragmentation at $E_{\rm ker} < 1$ eV, where they are inefficient [8,9].

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. Additionally, the He^+ ions also acquire a kinetic energy directly in the ionization process. In high-energy collisions, the momentum transfer $p_{\rm He^+}$ to the He^+ ions in an overwhelming majority of ionizing events does not exceed 1 a.u. [14,25,33] with the corresponding recoil energy of $2\times(p_{\rm He^+}^2/2M_{\rm He}) \lesssim 4$ meV. Therefore, so that the reflection approximation $E_{\rm ker} = 1/R$ may still be used, one must have $E_{\rm ker} \gg 4$ 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 $14 < R \lesssim 250$ Å (corresponding to $60 \text{ meV} \lesssim E_{\rm ker} < 1$ eV), where the dimer spends most of the time. Using the cross section for the production of two helium ions and the wave function of the dimer ground state, we can calculate the fragmentation cross section $d\sigma^{\rm frag}/dE_{\rm ker}$ representing the kinetic energy release spectrum [22]. In Fig. 5(a) we present it for collisions with 5 GeV/u U^{92+} projectiles.

The shape of the spectrum depends on the form of the dimer ground state and, hence, on its binding energy. The reported values for the binding energy I_b vary between 44.8 [34] and 161.7 neV [35], with 139.2 [36] and 151.9 neV [11] being regarded as most precise, having the relative difference of just 9%. However, in the range $0.06 \leq E_{\rm ker} \leq 1$ eV, these 9% are converted into 26% difference in the shape of the energy spectrum [see Fig. 5(b)]. Thus, due to the extremely long interaction range inherent to the ultrafast projectile, this spectrum spans a very broad diapason of R becoming sensitive to a small change in the dimer binding energy. This suggests that collisions with ultrafast projectiles can be used for determining this energy.

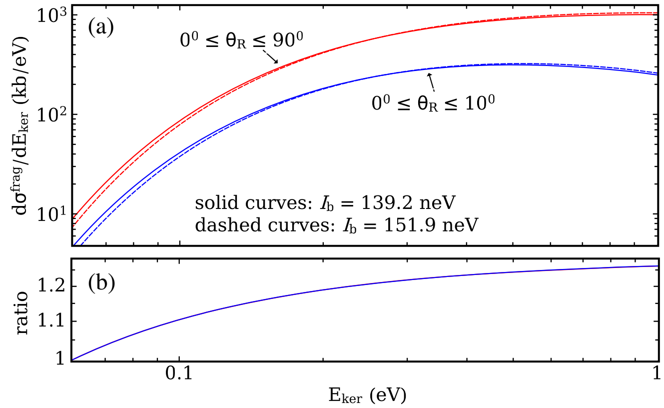

FIG. 5. (a) $d\sigma^{\rm frag}/dE_{\rm ker}$ for the fragmentation by 5 GeV/u U^{92+} averaged over the dimer orientation angle $\theta_{\boldsymbol{R}}$. (b) The ratio of the spectra obtained with $I_b = 151.9$ and 139.2 neV. The ratio was normalized to 1 at $E_{\rm ker} = 60$ meV and turned out to be practically the same for both orientation ranges.

According to [36], relativistic effects in free He_2 reduce I_b by about 14% resulting in ≈40% change in the shape of $d\sigma^{\rm frag}/dE_{\rm ker}$. However, this shape is much more affected by relativistic effects caused by very high impact energies. For instance, depending on the dimer orientation, these effects may enhance the lower-energy ($E_{\rm ker} \lesssim 0.1$ eV) part of the spectrum by 1–2 orders of magnitude.

In conclusion, we have studied the fragmentation of the helium dimer into singly charged ions by relativistic highly charged projectiles. It was found that the breakup events with kinetic energy release <1 eV 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 probe the structure of the dimer ground state in the halo region ∼(14–250) Å, where the dimer spends most of the time. A high sensitivity of the $E_{\rm ker}$ spectrum to the value of the dimer binding energy suggests that such projectiles can be used for its accurate determination.

Collisions with ultrafast projectiles can also be applied to explore the ground states of ^{6}LiHe and ^{7}LiHe dimers, which are other humongous diatomic molecules, having an average size of about 49 and 28 Å, respectively [37].

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[29] Note that the cross section for the He_2 fragmentation at $R = 500$ a.u. by relativistic U^{92+} projectiles are comparable in magnitude to those for the K-shell ionization of the heaviest atoms by the same U^{92+} projectiles.
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