

Paper III

8.3 Classical and quantum-mechanical investigation of the role of nondipole effects on the binding of a stripped HD^{2+} molecule

L. Sælen, I. Sundvor, T. Birkeland, S. Selstø, and M. Førre

Physical Review A, **76**, 013415 (2007)

Classical and quantum-mechanical investigation of the role of nondipole effects on the binding of a stripped HD²⁺ molecule

L. Sælen,¹ I. Sundvor,¹ T. Birkeland,² S. Selstø,^{1,*} and M. Førre¹

¹*Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway*

²*Department of Mathematics, University of Bergen, N-5007 Bergen, Norway*

(Received 25 January 2007; published 23 July 2007)

A quantum-mechanical approach is employed to study the binding of two bare nuclei of equal charge in strong laser fields beyond the dipole approximation [Smirnova *et al.*, Phys. Rev. Lett. **90**, 243001 (2003)]. The role of nondipole effects in the binding mechanism is investigated, and it is found that, in spite of a significant contribution to the dynamics, the nondipole effects do not alter the characteristic lifetime of the system. The results are supported by classical calculations addressing the question of decoupling of the center-of-mass and relative motions.

DOI: 10.1103/PhysRevA.76.013415

PACS number(s): 33.80.Rv, 33.80.Wz, 42.50.Hz

Quantum control, i.e., the ability to achieve a desired evolution of a quantum system by applying external fields, is a major theme of modern atomic, molecular, and optical sciences. From a theoretical point of view, quantum systems very quickly become incredibly difficult to handle with increasing complexity of the system. On the other hand, the dynamics of complex quantum systems can often be adequately described by quasiclassical models.

Recently, it was suggested [1], based on solutions of the time-dependent Schrödinger equation within the dipole approximation, that carefully selected combinations of strong linearly and circularly polarized laser fields could bind two same-sign charges, a proton and a deuteron. The nuclei are bound in the sense that the Coulomb explosion is significantly slowed down when the system is subject to external fields. Even though the cycle-averaged potential in this case does not give rise to stabilization, the mass difference of the particles gives an additional dynamical effect which could create a metastable state. Later, it was shown that the binding could in fact be described by classical mechanics [2], but that the spatial dependence of the laser fields may have a detrimental effect on the binding. However, care should be taken when the role of nondipole effects is investigated within a classical model [3], and a full quantum-mechanical analysis of the problem, with nondipole effects included, is important.

In this work, we reconsider the problem of binding of a proton and a deuteron with strong laser fields and discuss the role of nondipole effects in the binding mechanism. Supported by nondipole wave packet calculations, we find that, although the spatial dependence of the fields is substantial for the dynamics, it does not affect the degree of binding. In fact, by carefully selecting the initial wave function, the nuclei could apparently be even more strongly bound in the nondipole case than in the dipole situation. This illuminates how the initial wave packet influences the manifestation of the nondipole effects in the system.

We use atomic units (a.u.) with $m_e = e = \hbar = a_0 = 1$, except where specified otherwise. Our system consists of two par-

ticles with masses $m_{1,2}$, charges $q_{1,2}$, and positions $\mathbf{r}_{1,2}$, respectively. The nonrelativistic dynamics of two particles interacting with a classical electromagnetic field is governed by the time-dependent Schrödinger equation $i(\partial/\partial t)\Psi(\mathbf{r}_1, \mathbf{r}_2, t) = H\Psi(\mathbf{r}_1, \mathbf{r}_2, t)$, with the Hamiltonian

$$H = \frac{1}{2m_1}[\mathbf{p}_1 - q_1\mathbf{A}(\mathbf{r}_1, t)]^2 + \frac{1}{2m_2}[\mathbf{p}_2 - q_2\mathbf{A}(\mathbf{r}_2, t)]^2 + \frac{q_1q_2}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (1)$$

Following Smirnova *et al.* [1] and Madsen and Hansen [2], we use a linearly polarized field propagating in the x direction together with a circularly polarized field propagating in the z direction, corresponding to the vector potential

$$\mathbf{A}(x, z, t) = \frac{E_1}{\omega_1} \sin(\omega_1 t - k_1 z + \phi_1) \hat{x} + \frac{E_1}{\omega_1} \cos(\omega_1 t - k_1 z + \phi_1) \hat{y} + \frac{E_2}{\omega_2} \sin(\omega_2 t - k_2 z + \phi_2) \hat{z}. \quad (2)$$

We now expand the vector potential (2) to first order in the spatial derivatives and introduce center-of-mass (c.m.), $\mathbf{R} = (1/M)(m_1\mathbf{r}_1 + m_2\mathbf{r}_2)$, and relative coordinates, $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. Finally, we apply the Kramers-Henneberger (KH) transformations [4] $\Psi^{(KH)} = U\Psi = e^{i\alpha_R P} e^{i\alpha_r P} \Psi(\mathbf{r}, \mathbf{R}, t)$ on the wave function. Here,

$$\alpha_R(t) = -\frac{Q}{M} \int_0^t \mathbf{A}(\mathbf{0}, t') dt' \quad (3)$$

and

$$\alpha_r(t) = -\frac{\tilde{q}}{\mu} \int_0^t \mathbf{A}(\mathbf{0}, t') dt' \quad (4)$$

represent the positions relative to the laboratory frame of classical free particles with masses $M = m_1 + m_2$ (total mass) and $\mu = m_1 m_2 / M$ (reduced mass), and charges $Q = q_1 + q_2$ (total charge) and $\tilde{q} = \mu(q_1/m_1 - q_2/m_2)$ (reduced charge), respectively, moving in the dipole field. In the following, the vector potential $\mathbf{A} = \mathbf{A}(\mathbf{0}, t)$ and the electric field $\mathbf{E} = \mathbf{E}(\mathbf{0}, t)$

*Present address: Atomic Physics, Fysikum, Stockholm University, S-106-91 Stockholm, Sweden.

SÆLEN *et al.*

PHYSICAL REVIEW A **76**, 013415 (2007)

$= -\partial_t \mathbf{A}$ depend only on t . The Hamiltonian corresponding to the new point of view becomes

$$\begin{aligned} H^{(KH)} &= U H U^\dagger + i(\partial_t U) U^\dagger \\ &= H_R^{(KH)} + H_r^{(KH)} - \frac{\tilde{q}}{Mc} \mathbf{P} \cdot [\mathbf{E}^z(z + \boldsymbol{\alpha}_r \cdot \hat{z}) + \mathbf{E}^l(x + \boldsymbol{\alpha}_r \cdot \hat{x})] \\ &\quad - \frac{\tilde{q}}{\mu c} \mathbf{p} \cdot [\mathbf{E}^z(Z + \boldsymbol{\alpha}_R \cdot \hat{Z}) + \mathbf{E}^l(X + \boldsymbol{\alpha}_R \cdot \hat{X})], \end{aligned} \quad (5)$$

with

$$\begin{aligned} H_R^{(KH)} &= \frac{1}{2M} p^2 + \frac{1}{c} \left(\frac{Q^2}{M} + \frac{\tilde{q}^2}{\mu} \right) \mathbf{A} \cdot (\mathbf{E}^z Z + \mathbf{E}^l X) - \frac{Q}{Mc} \mathbf{P} \cdot [\mathbf{E}^z(Z \\ &\quad + \boldsymbol{\alpha}_R \cdot \hat{Z}) + \mathbf{E}^l(X + \boldsymbol{\alpha}_R \cdot \hat{X})], \end{aligned} \quad (6)$$

and

$$\begin{aligned} H_r^{(KH)} &= \frac{1}{2\mu} p^2 + \frac{q_1 q_2}{|\mathbf{r} + \boldsymbol{\alpha}|} + \frac{1}{c} \left(\frac{Q\tilde{q}}{M} + \frac{\tilde{q}q'}{\mu} \right) \mathbf{A} \cdot (\mathbf{E}^z z + \mathbf{E}^l x) \\ &\quad - \frac{q'}{\mu c} \mathbf{p} \cdot [\mathbf{E}^z(z + \boldsymbol{\alpha}_r \cdot \hat{z}) + \mathbf{E}^l(x + \boldsymbol{\alpha}_r \cdot \hat{x})]. \end{aligned} \quad (7)$$

\mathbf{E}^z and \mathbf{E}^l denote the circularly and linearly polarized parts of the electric field, $q' = (q_1 m_2^2 + q_2 m_1^2)/M^2$ is the effective charge [5], and $\mathbf{R} = [X, Y, Z]$ and $\mathbf{r} = [x, y, z]$. Higher-order terms proportional to $(1/c)^2$ along with purely time-dependent terms have been omitted.

As seen in Eq. (5), there are nondipole cross terms that prevent exact separation of the six-dimensional Schrödinger equation in the c.m. and relative coordinates. However, for the low-frequency and intense fields considered in this work, we can neglect nondipole terms of type $\mathbf{p} \cdot \mathbf{E}$ in Eqs. (5)–(7) compared to leading nondipole terms of type $\mathbf{A} \cdot \mathbf{E}$ for three reasons. First, the latter terms depend quadratically on the laser intensity [6]. Second, in contrast to the $\mathbf{p} \cdot \mathbf{E}$ terms, their magnitude is inversely proportional to the frequency of the applied field. Third, in the KH frame, and in the case of high-intensity fields, the particles essentially follow the motion of free particles in the field. Hence, the particles' momentum distributions are strongly centered about zero, making terms of type $\mathbf{p} \cdot \mathbf{E}$ even less significant [7].

Thus, by neglecting these terms, the Schrödinger equation separates and we are left with the following approximate Hamiltonian for the relative motion:

$$H^{(KH)} = \frac{1}{2\mu} p^2 + \frac{q_1 q_2}{|\mathbf{r} + \boldsymbol{\alpha}|} + \frac{1}{c} \left(\frac{Q\tilde{q}}{M} + \frac{\tilde{q}q'}{\mu} \right) \mathbf{A} \cdot (\mathbf{E}^z z + \mathbf{E}^l x). \quad (8)$$

We follow Smirnova *et al.* [1] and start with an initial Gaussian wave packet (in the KH frame)

$$\Psi_0 \propto e^{-(\alpha^2 + y^2)/\sigma_1^2} e^{-z^2/\sigma_2^2}, \quad (9)$$

with $\sigma_1 = 0.6$ and $\sigma_2 = 1.2$. The field parameters are $\omega_1 = 0.06$, $\omega_2 = 0.114$, $E_1 = 33$, and $E_2 = 260$. The nondipole effect of the linearly polarized field gives by far the dominant nondipole contribution to the dynamics. However, for completeness, the spatial dependence of the circularly polarized field

has also been included in the calculations. The three-dimensional Schrödinger equation with the Hamiltonians (7) and (8) is then solved numerically based on an accurate Fourier transform split operator method [8]. Converged results with an overall error of less than 2% were obtained with 180 grid points in the x and z directions, and 128 grid points in the y direction, with $-10 \leq x, y, z \leq 10$ a.u., and propagation time step $\Delta t = 0.036$ fs. An absorbing boundary is imposed at the edges.

Supported by numerical calculations, it is found that the Hamiltonians (7) and (8) yield practically identical results for the problem at hand. The difference is much less than 1% in all cases, suggesting that the approximation (8) is valid. This gives further evidence for the correctness of the decoupling of the six-dimensional Schrödinger equation, since the leading nondipole terms that prohibit separation in Eq. (5) are of the same order as the ones omitted in the approximation from (7) to (8).

We have also performed a classical trajectory Monte Carlo (CTMC) analysis of the six-dimensional problem and solved the Newtonian equations of motion for a large number of randomly picked phase-space points $(\mathbf{r}, \mathbf{p}, \mathbf{R}, \mathbf{P})$. The classical distributions resemble the probability density of the initial wave packet (9) in both the position and momentum space [2]. For the c.m. motion we use a distribution corresponding to $e^{-(x^2 + y^2)/\sigma_1'^2} e^{-z^2/\sigma_2'^2}$ with $\sigma_i' = \sqrt{\mu/M} \sigma_i$. Newton's equation of motion for two interacting charged particles in an electromagnetic field reads

$$m_i \ddot{\mathbf{r}}_i = q_i [\mathbf{E}(\mathbf{r}_i, t) + \dot{\mathbf{r}}_i \times \mathbf{B}(\mathbf{r}_i, t)] + (-1)^{i+1} q_1 q_2 \frac{\mathbf{r}}{|\mathbf{r}|^3}, \quad (10)$$

with $i = 1, 2$, and $\mathbf{B} = \nabla \times \mathbf{A}$ the magnetic field. In the dipole limit the equation for the relative motion becomes

$$\mu \ddot{\mathbf{r}} = \tilde{q} \mathbf{E} + q_1 q_2 \frac{\mathbf{r}}{|\mathbf{r}|^3}, \quad (11)$$

and in the KH frame, with $\mathbf{r}' = \mathbf{r} - \boldsymbol{\alpha}(t)$,

$$\mu \ddot{\mathbf{r}}' = q_1 q_2 \frac{\mathbf{r}' + \boldsymbol{\alpha}_r}{|\mathbf{r}' + \boldsymbol{\alpha}_r|^3}. \quad (12)$$

Going beyond the dipole approximation introduces a coupling to the c.m. motion, and we need to solve a six-dimensional problem. The CTMC calculations were done using 100 000 initial phase-space points, and the simulations were checked thoroughly for convergence. The classical equivalent of the Hamiltonian (8) was obtained directly from Hamilton's equations $\dot{x}_i = \partial \hat{H} / \partial p_i$, $\dot{p}_i = -\partial \hat{H} / \partial x_i$, and results were compared with the full classical solutions of the corresponding six-dimensional problem. In all calculations the results coincided within a few percent, showing that a first-order expansion of the fields in the spatial derivatives is indeed sufficient to describe the nondipole dynamics. Furthermore, the calculations confirmed that the decoupling of the c.m. and relative motions is valid. The problem is thus effectively reduced from six to three dimensions.

Madsen and Hansen [2] showed that classical mechanics can describe the binding of the two nuclei when the dipole

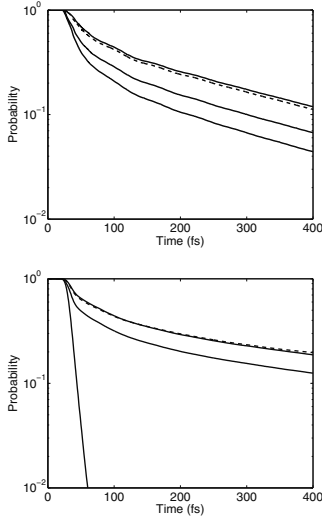


FIG. 1. Upper panel: Quantum-mechanical probability for the two particles to be less than $10a_0$ apart in the KH frame, and for three different combinations of the phases ϕ_1 and ϕ_2 . Solid lines: The solution with inclusion of retardation effects, and with $\{\phi_1, \phi_2\} = \{-\pi/2, \pi/3\}$ (upper line), $\{\phi_1, \phi_2\} = \{-\pi/2, \pi/5\}$ (intermediate line), and $\{\phi_1, \phi_2\} = \{0, 0\}$ (lower line). Dash-dotted line: The dipole result. Lower panel: Corresponding result of the classical CTMC calculations, with $\{\phi_1, \phi_2\} = \{-\pi/2, \pi/3\}$ (upper line), $\{\phi_1, \phi_2\} = \{-\pi/2, \pi/5\}$ (intermediate line), and $\{\phi_1, \phi_2\} = \{0, 0\}$ (lower line).

approximation is used, and obtained essentially the same result as in the quantum-mechanical approach [1]. However, when the full nondipole interaction was taken into account, it was found that trapping is less likely, from which they concluded that the spatial dependences of the fields have a detrimental effect on the binding. In Fig. 1 we show the probability as a function of time for the proton and deuteron to be less than $10a_0$ apart in the KH frame of reference. The upper panel presents the result of the nondipole *ab initio* wave packet calculations (solid lines) with the Hamiltonian (8), for three different combinations of the phases $\{\phi_1, \phi_2\}$. The dipole result (dashed line) is added for comparison. As expected, and due to the symmetry of the problem, the dipole result does not depend on the values of the phases. On the other hand, in the nondipole case this symmetry is broken. The lower panel shows the corresponding classical results, which indeed resemble and follow the quantum-mechanical results for all combinations of the phases ϕ_1 and ϕ_2 except one, i.e., $\phi_1 = \phi_2 = 0$. It turns out that the classical orbits are especially sensitive to nondipole effects for this particular choice of phases, and that the binding is significantly weakened. However, the quantum-mechanical results do not show the same degree of sensitivity, and reveal that the degree of binding is essentially the same, regardless of the values of

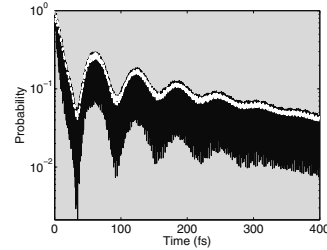


FIG. 2. Projection of the wave function on the initial wave packet (9) versus time for a case where the nondipole interaction is turned on for 5 fs, and for $\phi_1 = \phi_2 = 0$. The black and white curves show the results of the nondipole and dipole calculations, respectively.

the phases. But, more importantly, it was found that in the long term the decay is exponential in both the dipole and nondipole cases, with a common characteristic lifetime of $267(\pm 2\%)$ fs. Although a different choice of initial wave function than Eq. (9) or imposition of a laser pulse turn-on would strongly influence the short-term decay of the system, the exponential decay of the metastable component of the wave function is unaltered [1]. The facts that the classical results were extremely sensitive to the initial conditions and that the decay seemed to be nonexponential raise a question about the applicability of classical models in the nondipole regime [3].

Now, one could argue that, since the nondipole fields do not vanish at $t=0$ and vary with ϕ_1 and ϕ_2 , the actual differences in the results in Fig. 1, in effect, only reflect different choices of the initial wave function, and the observed nondipole effects are in some sense artificial. However, this is not

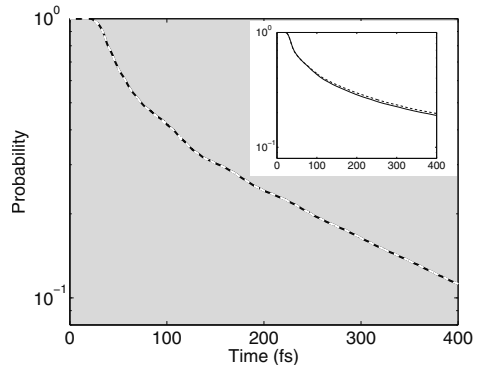


FIG. 3. Quantum-mechanical probability for the two particles to be less than $10a_0$ apart for the nondipole (dash-dotted line in black) and the dipole calculation (dash-dotted line in white), for the case in Fig. 2. The inset shows the classical result in the nondipole (solid line) and dipole (dashed line) limits.

SÆLEN *et al.*PHYSICAL REVIEW A **76**, 013415 (2007)

the case. Figure 2 shows the square of the autocorrelation function $|\langle \Psi_0 | \Psi(t) \rangle|^2$ as a function of time for a case where the nondipole terms are turned on slowly over the course of 5 fs. The turn-on amounts to starting the system at $t=5$ fs with an equally optimal initial wave packet for the nondipole case as is the wave function (9) for the dipole case. In this constructed example, the results of the dipole and nondipole calculations can be compared on equal footing. The clear difference between the dipole and nondipole results in Fig. 2 confirms that the nondipole terms have a significant dynamical effect. The very rapid oscillations of frequency $4\omega_2$ in the nondipole curve reflect the wave packet oscillations and are due to the dominating nondipole term in Eq. (8), namely, that of the linearly polarized field ($\sim \mathbf{A} \cdot \mathbf{E}$). Figure 3 shows the probability for the two nuclei to be less than $10a_0$ apart. In contradiction to the situation in Fig. 1, the phase dependence is now completely removed. With the turn-on, the degree of binding is identical in the dipole and nondipole cases, even in the short term, showing that the binding is not affected by the nondipole effects after all.

In conclusion, we have analyzed the problem of binding two bare nuclei, a proton and a deuteron, in strong laser fields, by solving the time-dependent Schrödinger equation for an initial Gaussian wave packet [1]. Our treatment has gone beyond the dipole approximation. We have shown that the binding of the stripped HD^{2+} molecule is maintained even when the nondipole interaction is taken into account. In fact, the decay of the initial wave packet is the same in the dipole and nondipole cases, illustrating that the binding is just as likely to occur in the nondipole limit. This opens the way for applications in controlling high-energy nuclear collisions on the femtosecond time scale [1]. We have also solved the problem within a quasiclassical model and found that the nondipole dynamics of the system at hand is not always adequately described by classical mechanics.

The present research has been supported by the Norwegian Research Council through the NANOMAT program and the Nordic Research Board NordForsk.

-
- [1] O. Smirnova, M. Spanner, and M. Ivanov, *Phys. Rev. Lett.* **90**, 243001 (2003).
 [2] L. B. Madsen and J. P. Hansen, *Phys. Rev. A* **71**, 045402 (2005).
 [3] M. Mahmoudia, Y. I. Salamin, and C. H. Keitel, *Phys. Rev. A* **72**, 033402 (2005).
 [4] W. Pauli and M. Fierz, *Nuovo Cimento* **15**, 167 (1938); H. A. Kramers, *Collected Scientific Papers* (North-Holland, Amsterdam, 1956), p. 866; W. C. Henneberger, *Phys. Rev. Lett.* **21**, 838 (1968).
 [5] H. R. Reiss, *Phys. Rev. A* **19**, 1140 (1979).
 [6] A. Bugacov, M. Pont, and R. Shakeshaft, *Phys. Rev. A* **48**, R4027 (1993).
 [7] J. R. Vázquez de Aldana, N. J. Kylstra, L. Roso, P. L. Knight, A. Patel, and R. A. Worthington, *Phys. Rev. A* **64**, 013411 (2001).
 [8] M. D. Feit, J. A. Fleck, Jr., and A. Steiger, *J. Comput. Phys.* **47**, 412 (1982).