Physics Letters A 373 (2009) 2556–2559

Contents lists available at ScienceDirect

Physics Letters A

www.elsevier.com/locate/pla

Rescattering and vibrations in homonuclear diatomic molecules in a strong electromagnetic field

Giuseppe Castiglia, Pietro Paolo Corso, Emilio Fiordilino *, Franco Persico

CNISM and Dipartimento di Scienze Fisiche e Astronomiche, Università di Palermo, Via Archirafi 36, 90123 Palermo, Italy

ARTICLE INFO

Article history:
Received 21 January 2009
Received in revised form 15 May 2009
Accepted 18 May 2009
Available online 21 May 2009
Communicated by P.R. Holland

PACS:
37.10.Vz
42.65.Re
34.80.Gs
33.15.Mt

ABSTRACT

The electron of a H_2^+ driven by a strong electromagnetic field induces molecular vibrations. Numerical and analytical results show that the molecule behaves as a parametric oscillator and can be treated as a kicked oscillator. The results are discussed from the point of view of the electron's periodic dressing and undressing processes.

Experimental results on the ultrafast nonlinear response of polarons (i.e. electrons surrounded by a cloud of virtual phonons) in *n*-doped GaAs have been recently reported [1] which are relevant for the fundamental process of undressing of particles in quantum field theory [2]. In this experiment an electron inside the semiconductor is accelerated by a high impulsive electric field in a time short on the scale of the period of lattice oscillations. Thus the electron shakes off the cloud of virtual phonons initially surrounding it and its dynamics inside the crystal suddenly changes from that of a dressed electron to that of a bare one. In addition, the virtual phonons initially belonging to the cloud surrounding the electron are suddenly transformed into a pulse of real phonons which are detected in the experiment. The inverse process, in which an initially bare electron in a semiconductor gets dressed by a cloud of virtual phonons, has been considered only theoretically in the past [3].

The aim of this Letter is to point out that, analogously to the polaron case, rapid (on the scale of attoseconds) and periodic (on the scale of femtoseconds) dressing and undressing events of an electron by phonons take place in homonuclear diatomic molecules subjected to a strong impulsive electromagnetic field, and to analyse the ensuing generation of real phonons in these structures which are more elementary than crystals and consequently lend themselves to a more accurate theoretical treatment. In this Letter we shall discuss this phenomenon entirely within a semiclassical framework, treating the electron quantum mechanically and the molecular vibrations classically, although (with a slight abuse of language) we shall often refer to these vibrations in terms of phonons. The extension of our theory to a quantum treatment of phonons will be the subject of further research. Here we only wish to anticipate that our physical model seems to lead, in a quantum framework, to the concept of the periodic polaron which, to the best of our knowledge, has never been considered previously and that in addition it seems to provide a simple experimental tool to investigate the dynamics of the kicked harmonic oscillator.

The electronic energy of a bound homonuclear diatomic molecule can be modelled as an effective potential function of the internuclear distance R. This potential has a minimum at an internuclear distance R_0, around which oscillations of R may take place. In aligned molecules the nuclear motion is essentially one-dimensional. In the absence of external forces and for small vibration amplitudes this motion can be considered harmonic. Thus the effective potential of the system can be approximated as [4]

$$V(R) = V_0 + \frac{1}{2}k(R - R_0)^2 \tag{1}$$

where k is the interatomic force constant. The parameters V_0, k and R_0 are influenced by the configuration of the electrons in the molecule. Thus a sudden change of this electronic configuration is expected to yield a sudden change in these parameters, presumably accompanied by the onset of molecular oscillations.

Suppose now the diatomic molecule is subjected to a strong electromagnetic pulse. Then, according to recollision theory [5,6] and using the single active electron ansatz, a small portion of the electronic wavefunction, in the form of an electronic wavepacket,

---

[*] Corresponding author.
E-mail address: fiordili@fisica.unipa.it (E. Fiordilino).

0375-9601/$ – see front matter
doi:10.1016/j.physleta.2009.05.028

should periodically abandon the molecule and return to it after half a laser cycle [7]. The duration of each abandonement and reabsorption act is expected to last a fraction of the laser period according to the ADK theory [8], equivalent to a time interval of the order of ≈ 100 as. This is short enough on the molecular vibration scale to be considered as a sudden, albeit small, change in the electronic density. As a result, the molecule is subjected to a periodic shake-up, whose effects should become manifest in the vibrational dynamics of the molecule.

In principle, this periodic shake-up could be due to the impulsive change of any of the parameters V_0, k and R_0. In this Letter we shall focus on the case in which the effect of the impulsive change is mainly felt by k. For this case we shall produce approximate analytical results, which we shall compare with those of an exact numerical treatment of the very special diatomic molecule H_2^+. This particular choice is motivated by previously published theoretical evidence that the HHG spectrum of H_2^+ is sensitive to molecular vibrations [9,10], although also other molecules have been found whose dynamics in strong fields is affected by vibrational motion [11]. As we shall see, reasonable agreement will be obtained between our analytical and numerical treatments. This agreement will lead us to conclude that H_2^+ in a strong electromagnetic field can be approximately regarded as a simple and flexible experimental realization of the kicked harmonic oscillator. An important additional reason for the choice of H_2^+ is that it is the simplest possible diatomic molecule, amenable to a satisfactory mathematical treatment without the necessity to deal with electron–electron interaction. Needless to say, the drawback of this choice is that a classical treatment of the nuclear motion in the case of H_2^+ may be considered questionable in view of the relatively small proton mass. Nevertheless the physically transparent results obtained seem to suggest that the main physics of our model can be extended to heavier homonuclear diatomic molecules, where a classical treatment of the nuclei motion is expected to be valid.

The numerical model we adopt here is the same one-dimensional model presented in [12] and used in [9]. Thus the electron (mass m, charge $-e$) dynamics is described by the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi(x,t) = \left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2} + V\big(R_1(t), R_2(t); x\big) + ex\mathcal{E}(t)\right]\psi(x,t) \tag{2}$$

where x is the electron coordinate and $\mathcal{E}(t)$ is the laser driving field. R_1 and R_2 are the nuclei positions, whose time-dependence is governed by the classical Newton equations

$$M\frac{d^2}{dt^2}R_i = F_{ie}(t) + F_{ij}(t) + e\mathcal{E}(t), \quad i \neq j = 1, 2. \tag{3}$$

In this equations M is the proton mass. Moreover in Eq. (2)

$$V(R_1, R_2; x) = -\frac{e^2}{\sqrt{A + [x - R_1]^2}} - \frac{e^2}{\sqrt{A + [x - R_2]^2}} \tag{4}$$

where A is an appropriate screening parameter. F_{ie} is the attractive force exerted by the electron cloud on the ith nucleus

$$F_{ie}(t) = -\int \frac{e^2[x - R_i]|\psi(x,t)|^2}{\{A + [x - R_i]^2\}^{3/2}}dx, \quad i = 1, 2, \tag{5}$$

which is a functional of the electron density in the neighborhood of the molecule and F_{ij} is the repulsive internuclear force of the Coulomb form with a screening parameter A_N. In the absence of $\mathcal{E}(t)$ for $A = 0.5$, $A_N = 0.03$ and for constant R_i, the ground state energy of the molecule is minimum for $R(t) = |R_1 - R_2| = R_0 \approx 1.935$ a.u. We take this as the initial configuration of the system and at $t = 0$ we switch suddenly on a sinusoidal field $\mathcal{E}(t) = \mathcal{E}_0 \sin(\omega_L t)$.

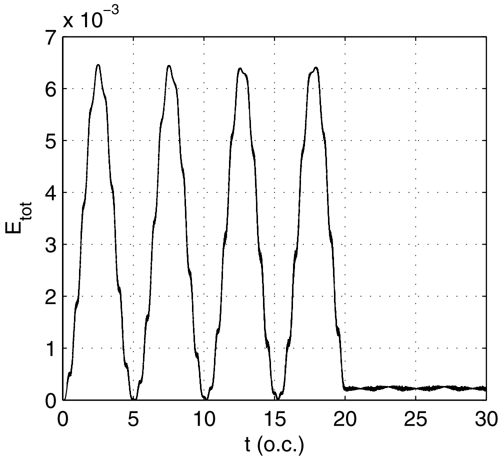

Fig. 1. The vibrational energy E_{tot} absorbed by the molecular ion as function of time t in optical cycles. The laser field is switched off at $t = 20$ o.c.

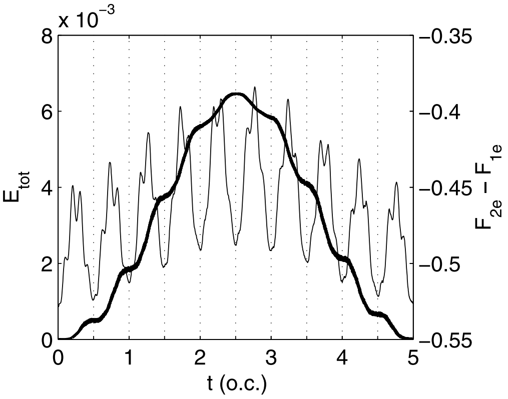

Fig. 2. Thick line: magnification of the first part of Fig. 1; it displays more clearly the bumps and shows more detail, such as their duration and location. Thin line: plot of the force $F_{2e} - F_{1e}$ exerted by the laser driven electron upon the nuclei. The bumps are in temporal coincidence with the minima of the force.

In Fig. 1 we plot the time evolution of the vibrational energy transmitted to the ion by the laser at time t. This energy is obtained numerically by solving the coupled equations (2) and (3) by means of a split-operator code, using a 780 nm laser pulse whose intensity is 5×10^{14} W/cm^2. At $t = 20$ o.c. (optical cycles) the laser field is switched off suddenly and the molecule vibrates freely. It should be noted that, perhaps nontrivially, for $t < 20$ o.c. this energy is a periodic function of time at frequency close to $\omega_0 = \sqrt{2k/M}$. In our model the small additional bumps of period half laser cycle, shown more clearly in Fig. 2, are located at the instants of departure and return of the rescattered electronic wavepacket responsible also for the emission of the spectrum. These bumps are expected on the basis of the physical argument presented above and of the form of (5), which suggests that at these instants $F_{ie}(t)$ changes within a time interval of the order of ≈ 100 as; in Fig. 2 we show also the total force $F_e(t) \equiv F_{2e}(t) - F_{1e}(t)$ exerted by the rescattered electron upon the nuclei.

The main oscillations at frequency $\simeq \omega_0$ can be understood by the following argument.

The sudden increases and decreases in $F_e(t)$ introduce a time dependence in k. We shall tentatively assume this time depen-