LETTER

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Theoretical study of attosecond laser interference on radioactive decay of cesium-137

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Abstract – At present, the research on reducing radioactive contamination has special significance. Professor Sugihara found that the $\langle H^+, e^- \rangle$ fragments in the plasma could reduce cesium-137 radiation activity. In this paper, a well-established method based on direct numerical solution of the three-dimensional Schrödinger equation is applied to study the interaction between the cesium-137 nucleus and the plasma. It is found that the plasma is completely formed in the first three optical periods of 800 nm laser. From the fourth optical period, the electrons that generated from the decay of cesium-137 become the main source of electrons in the plasma. In order to maintain the stability of the plasma, cesium-137 has to accelerate its decay within a certain time. The intrinsic mechanism of the rapid reduction of the radiation activity of cesium-137 was explained by theoretical calculation for the first time.

In recent years, the reduction of radioactive nuclide activity and its related research has become a hot topic in scientific research. It is found that the interaction between the ionized electrons and hydrogen ions does not lead to the formation of hydronium ions and hydroxyl radicals, but generates the stable plasma of $\langle H^+, e^- \rangle$ in water. Its potential is named as an extended spin-1 particle "infoton", which vibrates radially while rotating around the axis [1,2]. Professor Sugihara's discovery has aroused interest in the study of how plasma interferes with the decay of radio nuclides.

With the rapid development of attosecond science, the one-dimensional time-dependent wave packet method can be used to explain the coherent superposition mechanism of electron and nuclear wave packets [3]. Selstø *et al.* used a three-dimensional time-dependent method to calculate the ionization in the attosecond high frequency and intense field. It is shown that the ionization probability was related to both the nuclear distance and the orientation. The orientation between the laser field and the molecular axis in space can affect the generation of higher harmonics [4]. As a coherent wave packet method is developed by Corkum *et al.*, a large number of experiments on generation of higher harmonics in gas and solid states have been conducted [5–7]. For liquid state, harmonic absorption of liquid membrane is inevitable. With the emergence of liquid plane-micro-jet technology, liquid planar film technology has developed rapidly [8,9]. The microscopic planar jet designed by Luu *et al.* can reduce the film thickness to less than 2 μm [10]. Thus the harmonic absorption of liquid films can be effectively weakened. When the target is subjected to an intense laser field, a cloud of evaporating gas quickly forms around the liquid target. The liquid film is designed as wedge structure to make full use of the refractive index of wavelength dependence, which can naturally dissociate the higher harmonics generated by the liquid from those by the surrounding gas in the far field of space, so as to realize the XUV band observation of liquid higher harmonic radiation in experiment [10]. Therefore, it is possible to apply attosecond laser technology to liquid water to track the plasma formation process [11–15].

In this paper, a laser with intensity of 10^{14} W/cm^2 is applied, and the time-dependent properties of the laser pulse are used to track the above collision, recombination and plasma formation processes. Firstly, the probe pulse weakens the binding force between the electron and neutral H_2 molecule, then one of the bonding electrons becomes free, and the vibrational wave of H_2^+ ion launches on

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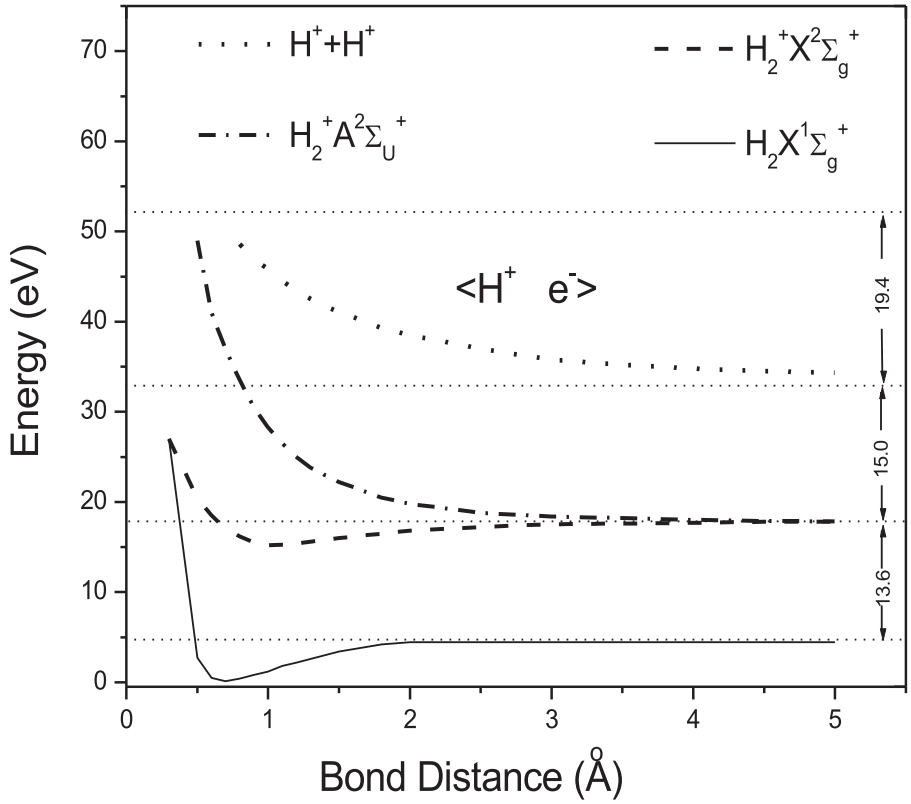

Fig. 1: The potentials of H_2 and H_2^+ during our calculation are shown. The solid line represents the ground state of hydrogen molecule; the dashed line represents $H_{2^+}(X_g^{2^+})$ state; the dash-dotted line represents $H_{2^+}(A_U^{2^+})$ state; the dotted line represents double ionization state.

its ion surface $H_2^+(X^2\Sigma_g^+)$ immediately [16]. Meanwhile, the electronic and vibrational wave packets are correlated within the time scale of the laser oscillation. During the first three optical periods the ionized electron will return several times to re-collide with its parent ion, which leads to a transition of ion density to excited state, where double ionization is generated with the concentration of a large number of H^+ ions, as shown in fig. 1.

The effect of the intense laser field not only provides a means to generate plasma, but also makes it possible to track the interaction between the $\langle H^+, e^-\rangle$ plasma and the radioactive nucleus in real time. The stability of the plasma is based on H^+ ion concentration that is caused by the dissociation and double ionization. Moreover, with the recombination between H^+ ions and electrons, high harmonic signals are generated, which are presumed to be the mechanism of terahertz radiation energy releasing [17].

If a soil or food sample containing cesium-137 is immersed in an aqueous solution used as a liquid target, and an intense laser field is directly applied around the target by hydrogen vapor generated by hydrolysis, the time-dependent wave packet equation for the coherent motion of the electrons and the nuclei can be established [18–23].

In view of the time scale, the electronic wave function becomes stationary in 0.9 fs during a field-induced transition, which is different from that of the nucleus. It is not until the time increases above 4 fs that the wave function of the nucleus motion begins to change appreciably, which makes the density of the nucleus unstable. This situation continues until 60 fs later, and the density of the nucleus basically remains unchanged and reaches a stable state.

The three-dimensional (3D) time-dependent wave packet method, *i.e.*, the $\langle H^+, e^-\rangle$ plasma in 2 dimensions and the nucleus of cesium-137 in 1 dimension has been carried out to calculate the re-collision between the plasma and the nucleus, shown by eq. (1). The quantum coherence problem between the electrons and the $\langle H^+, e^-\rangle$ plasma can be calculated by eq. (2). Equation (3) is the motion equation of H, whose initial wave function is the ground state of H,

$$i\hbar\frac{\partial}{\partial t}\psi_I(R_{Cs},\vec{R},t)=\left[-\frac{\hbar^2}{2\mu_1}\nabla^2+V_I\right]\psi_I(R_{Cs},\vec{R},t), \quad (1)$$

$$i\hbar\frac{\partial}{\partial t}\psi_{II}(R,\vec{r},t)=\left[-\frac{\hbar^2}{2\mu_2}\nabla^2+V_{II}\right]\psi_{II}(R,\vec{r},t), \quad (2)$$

$$i\hbar\frac{\partial}{\partial t}\psi_{III}(R_H,t)=\left[-\frac{\hbar^2}{m_H}\frac{\partial^2}{\partial {R_H}^2}+V(R_H,t)\right]\psi_{III}(R_H,t). \quad (3)$$

In the above equations, $\vec{R}$ is the space coordinate vector of the stable constituent electron in the plasma, $\vec{r}$ is the

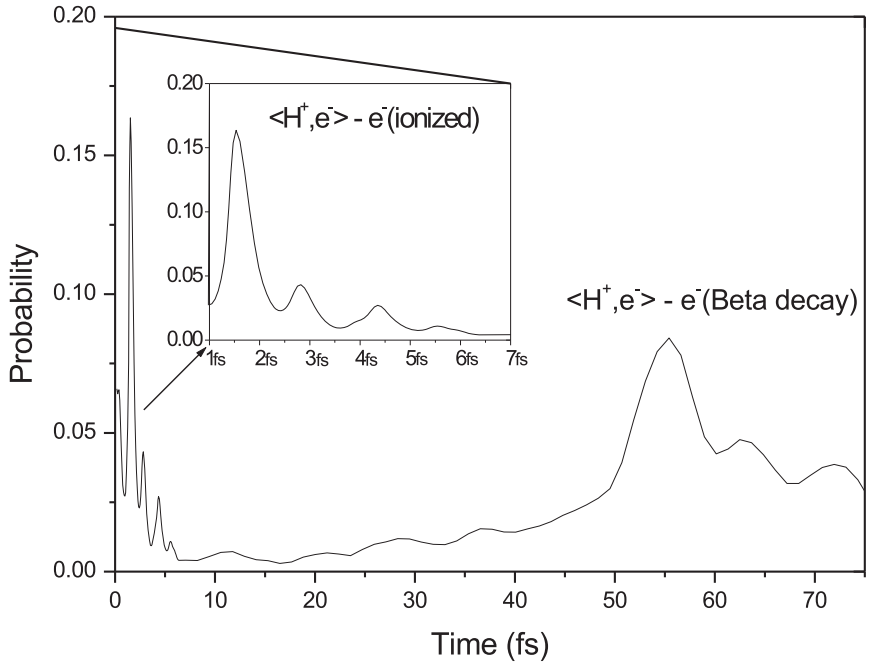

Fig. 2: The time-dependent distribution of electron-plasma collision probability is shown. The distribution of collision probability within the first three optical periods of 800 nm wavelength laser is shown in the inset.

space coordinate vector of an unstable electron with multiple return and collision after ionization. Furthermore, R_{Cs} and R_H are the coordinates of cesium-137 and H, respectively, μ_1 is the reduced mass of the stable constituent electron and the cesium-137 nucleus, μ_2 is the reduced mass of the stable constituent electron and the unstable ionized electron.

The interaction potential between the plasma and cesium-137 can be denoted by $V_{\rm I}$:

$$V_{\rm I} = V_{II} + V_K, \tag{4}$$

$$V_{II} = V(R_H, t) + V_C + V_\Gamma + V_R, \tag{5}$$

where $V(R_H, t)$ denotes the interactions between bound electrons and H nuclei, which include the ground state ($H_2(X^1\Sigma_g)$), the ionic states ($H_2^+(X^2\Sigma_g^+)$, $H_2^+(A^2\Sigma_u^+)$), and the double ionic state, which is extended to the continuum ionic states. The Coulomb interaction between the emitted electron and the nuclei V_C is added to the diagonal items of the continuum ionic states. $V_\Gamma = -2\Gamma\hbar\frac{\partial^2}{\partial t^2}$, being associated with the real relaxation time, is recognized as a kind of frictional force to describe the interactions between the active electron (ionized and decayed) and its surroundings, which are added to the off-diagonal items of the continuum ionic states [24] (m_e, α and c denote one electron mass, fine structure constant and the speed of light in vacuum, respectively). V_R denotes the interactions among the nuclei, which are weak enough to be ignored. $V_K = -G\mu_{H^+ + e^-}M_{Cs}K(p)$ is the attractive potential between the $\langle H^+, e^-\rangle$ plasma and the nucleus of cesium-137, whose force range is very short (about picometers scale), where $K(p) = \lim_{r\to 0}\frac{3}{\pi}(\frac{2\pi r - L_p(r)}{r^3})$.

Equations (1) and (2) can be calculated by the time-dependent Schrödinger equation with the split-operator method. Solving the Schrödinger equation in 3 dimensions will increase the accuracy of the energy spectra for both the plasma and the nuclei. Figure 2 shows the probabilities of re-collision between the electrons and the $\langle H^+, e^-\rangle$ plasma with 800 nm wavelength. High-frequency collisions occurred in the first three optical periods, with the duration of each collision being about 1 to 2 fs, and the maximum probability of collision occurred in the first optical period. At the end of the third optical period, the collision probability decreased to a very low level. From the fourth optical period, the collision probability showed a steady increasing trend, and high probabilities occurred between 50 and 60 fs. It is inferred that the 8 femtoseconds (within the first three optical periods of 800 nm wavelength laser) is the plasma formation period, during which the additional H^+ ions mainly come from the collisions between the ionized electrons and H_2^+ ions. From the beginning of the fourth optical period, the plasma is mainly distributed on the repulsive potential. The dynamic recombination processes between H^+ ions and the ionized electrons are significantly enhanced, which consumes the ionized electrons and radiates energy in a high harmonic form. Over time, either the dissociation of H_2^+ ions or the collisions between the ionized electrons and the H_2^+ ions will temporarily form the H^+ ions clusters far more numerous than the number of the ionized electrons. At this point, the number of the electrons in the plasma should be significantly less than that of the H^+ ions, and the plasma appears to be temporarily unstable. The strong stability of these H^+ ions makes the plasma need a large number

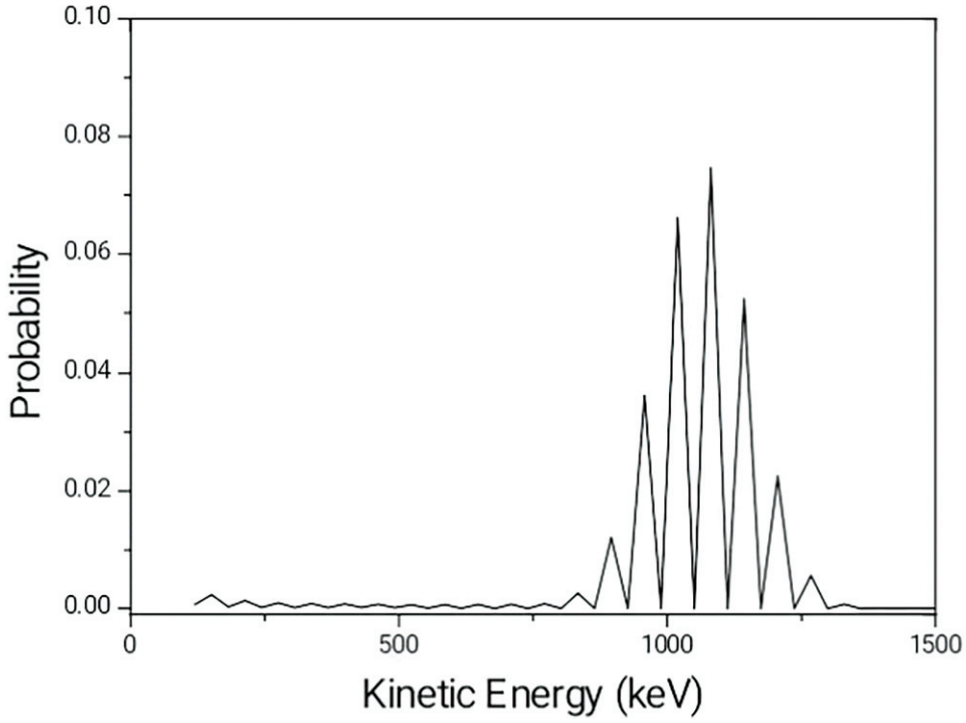

Fig. 3: The distribution of collision probability (between the plasma and cesium-137 nucleus) is shown.

of freely moving electrons as a supplement to maintain its relative stability.

However, under the action of the attractive potential, the electrons generated by the radioactive decay of the cesium-137 nucleus will approach and collide with the plasma, so the cesium-137 nuclei must emit beta rays more quickly to compensate for the loss of the ionized electrons and thus maintain the stability of the plasma.

The collision between the $\langle H^+, e^- \rangle$ plasma and cesium-137 should have occurred during the formation of the plasma. Although the kinetic energy of the plasma is low the collisions are inevitable, which is consistent with the results of Belyaev *et al.* [25]. Since the time-dependent Schrödinger equation contains the coupling between the $\langle H^+, e^- \rangle$ plasma and cesium-137, the energy distribution of the plasma naturally includes the relevant information about the motion of the cesium nucleus. The distribution of collision probability between cesium-137 and the plasma as a function of kinetic energy is shown in fig. 3. When the kinetic energy of $\langle H^+, e^- \rangle$ plasma is close to 1000 keV the collision probability increases significantly. The maximum collision probability occurs between 1100 keV and 1200 keV, which is close to the kinetic energy of the "infoton" particle proposed by Professor Sugihara [1,2]. The probability distribution of high frequency collisions between 1100 and 1400 keV indicates that the plasma is indeed stable and has frequent collisions with the nucleus of cesium-137.

In this paper, the three-dimensional Schrödinger equation has been applied to track the formation, development and stability of the plasma. Compared with ionized electrons, the strong stability of H^+ ions makes the electric neutrality of plasma need other electrons as a supplement. Cesium-137 will inevitably accelerate its decay in a certain time, release electrons, and contribute to maintaining the stability of the plasma. This is the fundamental reason why the "infoton particle" proposed by Professor Sugihara will rapidly reduce the decay activity of cesium-137 [1,2]. It is reasonable to believe that in the near future this work will pave the way for the emergence of new methods to reduce radioactive contamination.

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Data availability statement: All data that support the findings of this study are included within the article (and any supplementary files).

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