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Formation of the XeI excimer sustained by dielectric barrier discharge in Xe/I_2 at low pressure*

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Dielectric barrier discharge is used to study the mechanism of XeI excimer formation in the mixture of Xe and I_2 at low pressures (<1330 Pa). Fluorescence emission in the spectral region of 200–260 nm is examined. We report on the characteristics of the 253 nm emission intensity which varied with different total pressure. The results indicate that under the present experimental conditions, electron impact is the major reaction producing the excimer XeI(B), interpreted as $Xe^+ + I_2^- \rightarrow XeI(B) + I^*$, then radiating 253 nm fluorescence from transition B→X. The 253 nm emission increases with the total gas pressure up to a maximum value at a pressure of about 540 Pa, then decreases as the gas pressure is further increased. The 206 nm emission is determined by I^* from ionic recombination between Xe^+ and I_2^-. This result differs from previous works under other experimental conditions.

Keywords: Dielectric barrier discharge plasma, XeI excimer, fluorescence emission spectrum
PACC: 3350, 5280, 5220F

1. Introduction

In the past decade, considerable attention has been focused on the application of photons to the initiation of new chemical reactions or excited molecules.[1] Most of the photo-induced processes have been conducted with narrow wavelength excimer lasers. Many papers have been published on the mechanism of the excimer,[2,3] indicating that appropriate excimer laser media can provide an enormous potential for the development of lasers. The XeI excimer is the very candidate. It is a dimeric molecule in an excited state. Under normal physical conditions, the XeI excimer does not possess a stable ground electronic state; and once formed, it decomposes within a few nanoseconds, giving up its excitation energy in the form of vacuum ultraviolet (VUV) or UV photons. This makes the XeI excimer an extremely useful candidate as a very efficient light source since there can be no self-absorption or radiation trap resulting from transitions to and from the ground state. On the other hand, the XeI excimer has a continuous radiation spectrum that takes advantage of the laser adjustment of wavelength. The lifetime of the ground state is so short that it is of great benefit for high-frequency operations. Additionally, the binding energy of rare-gas iodide is expected to be smaller than that of the lighter halide-rare-gas complexes since iodine has the smallest electronegativity. Thus many sources are expected to be capable of being more efficient.

Previous research on the XeI excimer has mostly focused on high-pressure (>13300Pa) excimers.[4,5] However, a few works on the mechanisms of XeI formation at low pressures have been reported. In this paper we investigate the kinetics and radiative processes leading to UV emission from XeI excimers in the mixture of Xe/I_2 in a dielectric barrier discharge (DBD) plasma at low pressures (70–1330Pa). We have found that in this pressure range XeI(B) is likely to be formed by a reaction between Xe^+ and I_2^-, namely $Xe^+ + I_2^- \rightarrow XeI(B) + I^*$. This conclusion differs from previous works conducted at high pressures, as well as at low pressures under other experimental conditions.

2. The formation of the XeI excimer

The reaction mechanism of the XeI excimer is rather complicated, involving several atomic and molecular species in the ground state, several ionic species, and large numbers of excited atomic and molecular species.

Several key reactions for the formation of XeI excimers can be described as follows.[5]

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The high-energy electrons ionize and excite the xenon and iodine species in the reactions

$$e^- + Xe \rightarrow Xe^* + e^-,$$

$$e^- + Xe \rightarrow Xe^+ + 2e^-,$$

$$e^- + I_2 \rightarrow I + I^-.$$

At high pressures the XeI^* excimers can be created by a three-body ionic recombination or the harpoon reaction[6] in which the excited xenon species transfers its loosely bound electron to the iodine molecule, to form an electronically excited state of XeI^*

$$Xe^+ + I^- + M \rightarrow XeI^* + M,$$

$$Xe^+ + I_2^- \rightarrow XeI^* + I.$$

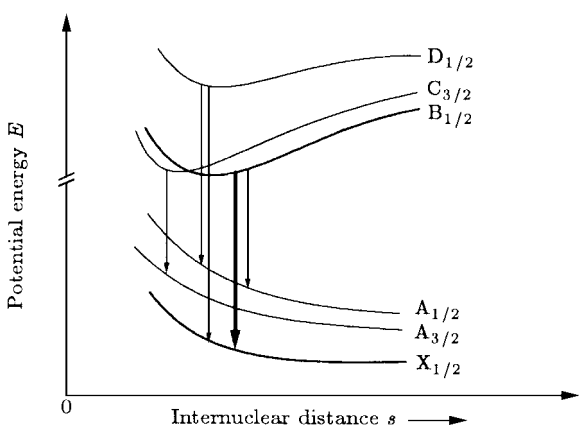


Fig.1. A diagram of the simplified potential energy scheme of XeI.

A simplified potential energy diagram of the xenon iodide excimer is shown in Fig.1. The ground state is only weakly bound and correlated with the ground state ^1S xenon and ^2P iodine. At a close internuclear separation, the low-lying ^2P states are split into, for example, $X_{1/2}$, $A_{1/2}$, and $A_{3/2}$ states. The three close-lying excited states, $B_{1/2}$, $C_{3/2}$ and $D_{1/2}$ as indicated, are generated by a positive xenon ion ^2P and a negative iodine molecular ion ^1S for a much larger internuclear separation. There are different transitions from the excited states and to the ground states, namely: $B_{1/2} \rightarrow X_{1/2}$, $B_{1/2} \rightarrow A_{1/2}$, $C_{3/2} \rightarrow A_{3/2}$, and $D_{1/2} \rightarrow X_{1/2}$, which correspond to different intensities.

The excimers are not very stable and rapidly decompose within a few nanoseconds, radiating UV photons, according to

$$XeI^* \rightarrow Xe + I + h\nu.$$

Additionally, this radiation process competes with several quenching reactions.

3. Experiment

Figure 2 shows a schematic diagram of the experimental apparatus. The discharge is sustained in a cylindrical glass tube, 5 mm in diameter and 140 mm in length, whose electrodes lie on two sides of the discharge tube. One electrode is overlaid with a layer of common glass. The UV fluorescence is output through a quartz slice of 2 mm in thickness, which is the unique window of the discharge tube. For the recording of emission spectrum we use an AM-566 grating monochromator (made in ARC., USA) in conjunction with a photomultiplier tube. The discharge is supplied by 30 kHz ac power with a voltage of 5 kV.

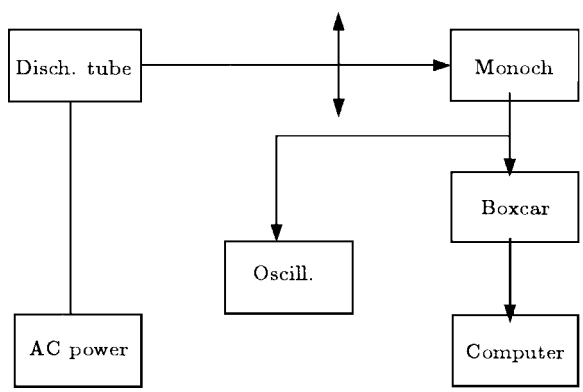


Fig.2. Schematic diagram of the experimental apparatus.

In the experiment, saturate iodine vapour is injected into the discharge tube first, and then mixed with Xe at different pressures. When the discharge plasma becomes steady, the fluorescence can be focused on the entrance slit of the monochromator, and input to a Boxcar followed by photomultiplier transfer. Finally, a computer records the spectrum. At the same time, an SR37 oscillator is used to monitor the total process.

4. Results and discussion

In the range of 200–260 nm, the emission spectrum from the mixture of xenon and iodine at a pressure of 70–1330 Pa is obtained in the DBD plasma. The spectrum shown in Fig.3 is observed at a current of 30 mA, and a total pressure of 540 Pa.

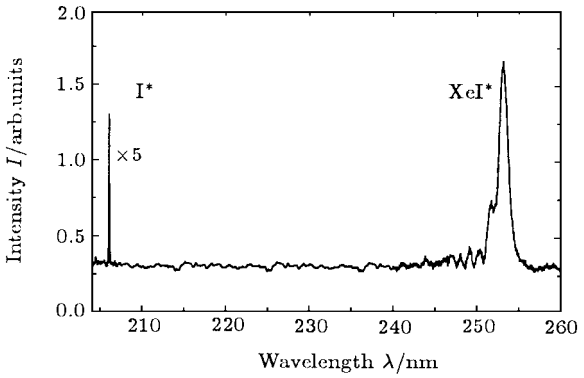


Fig.3. Fluorescence emission spectra at 540 Pa.

The intensity of the sharp-peaked emission at 253 nm appears to be strong in this spectrum. This band is not observed in the emission spectrum of the pure iodine or pure xenon. As far as the width is concerned, it is about 1.3 nm and unlikely to be of an atomic emission, nor the bound–bound transition of a common molecule according to the Frank–Condon principle.[7] So this band should be attributed to the bound–free transition corresponding to the transition $B_{1/2} \rightarrow X_{1/2}$ of XeI* according to the potential curve provided by Barns and Kushner[8] Otherwise, the undulatory structure is typical for this dispersion band, which is considered to be transitions from different vibrational levels to the ground state. The strongest peak lies at the red end of the total structure.

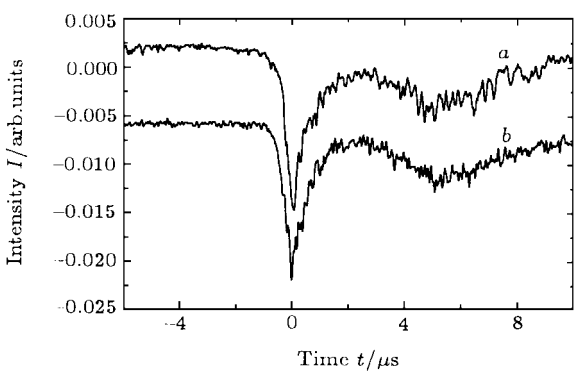


Fig.4. Time resolution diagram of I* and XeI*.

The atomic iodine emission line at 206 nm also appears in this spectrum, and the spectrum line corresponds to the transition $^2P_{3/2} \rightarrow {}^2P_{1/2}$.[9] The intensity of this line is stronger by far than that generated by the discharge plasma in pure iodine, and is directly proportional to the intensity of 253 nm emission varied with the pressure of xenon. Therefore, the appearance of the iodine emission line is related nearly to the participant of xenon. In order to explain this mechanism, we measure the temporal characteristic curves of the 206 and 253 nm emissions at the same discharge period. The time resolution diagram is shown in Fig.4. Curve *a*, which is of the 206 nm emission of I*, is enlarged five times in order to compare it easily with curve *b* which is of the 253 nm emission of the XeI* excimer. This figure shows that the signals of the 206 and 253 nm lines are generated simultaneously, and reach the maximum at the same time. So I* and XeI* are likely to be generated in the same reaction.

Under this condition, the electron temperature in the discharge plasma is about 21.5 eV, which is 1.5–2 times that of the first ionization potential of xenon atoms and goes beyond the ionization potential of iodine molecules. Therefore, there are a great deal of xenon ions and iodine molecular ions in the DBD plasma. They interact with each other to produce the excited state of the XeI excimer. As a result the main channel of the formation of the XeI excimer in the DBD plasma can be described as follows. Impacted by the high-velocity electrons, the atoms and molecules in the plasma are ionized to produce Xe^+ and I_2^-, then they combine with each other to generate the B excited state XeI excimers and excited iodine atoms, namely $Xe^+ + I_2^- \rightarrow XeI(B) + I^*$. XeI(B) and I* radiate the fluorescence of 253 and 206 nm respectively, when they transit to the lower levels.

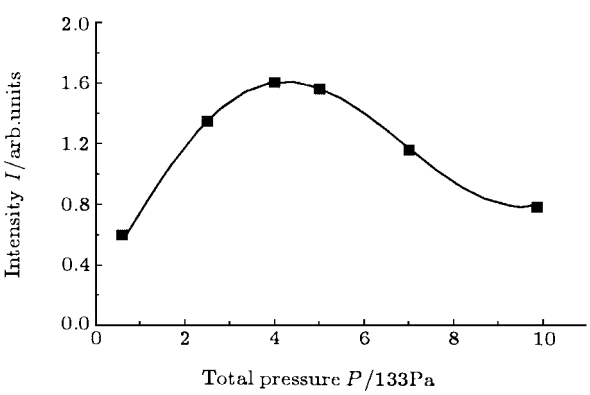


Fig.5. Intensity of 253 nm emission as a function of gas pressure.

At low pressures, the intensity of 253 nm emission as a function of gas pressure is shown in Fig.5. In the range of 70–1330 Pa, the 253 nm emission ($B_{1/2} \rightarrow X_{1/2}$) intensity increases with the total gas pressure up to the maximum at a pressure of about 540 Pa, but decreases with further increasing gas pressure. For the front part of the curve, with the increasing pressure of xenon, the density of ionic xenon increases, so the quantity of XeI* excimers produced increases.

For the part of the curve after the maximum, there are mainly two points to be stated. Firstly, the formation of Xe_2I^* with the increasing density of xenon replaces that of XeI^*.[10] Secondly, the ionization efficiency of the xenon species decreases while the time of interaction between the high-velocity electron and xenon reduces with increasing electron temperature.

5. Conclusion

Through the study of the emission intensity of XeI excimers in the DBD plasma at low pressures, we have found that the XeI(B) excimer can be generated by a DBD in the mixture of xenon and iodine. It then transits to the ground state with radiating the 253 nm fluorescence, and the major source of the XeI(B) excimer comes from the reaction $Xe^+ + I_2^- \rightarrow XeI(B) + I^*$. The 253 nm emission intensity increases with an increase of the total pressure in the range of 70–540 Pa, but its intensity decreases with a further increase of gas pressure. Additionally, the 206 nm emission generated simultaneously comes from I^* generated through ionic recombination between Xe^+ and I_2^-.

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