Depth-resolved dynamics of ion-tracks: correlation between VUV- and VIS-excimer luminescences from near-liquid and liquid helium irradiated with 4 MeV/amu N- and He-ions

Kazuie Kimura
Institute of Physical and Chemical Research (RIKEN), Wako Saitama 351, Japan

Track-depth resolved VUV (vacuum ultraviolet)-luminescence due to the transition $^1A^{-1}X$ has been measured on a dense helium target with a varying helium density. With increasing excitation density, the formation of $^1A$ is enhanced by the $^3a+^1a$ reaction and suppressed by the $^1a+^3a+^1a$ reaction. An additional peak in luminescence efficiency $dL/dE$ is much more intense than that for VIS (visible) $dL/dE$. The peak is caused by charge exchange and direct excitation processes.

1. Introduction

One of the most characteristic radiation effects of heavy ions in condensed matter is the high density excitation of electrons in the outermost shell. Additional effects such as charge-exchange and direct excitation may become important in the region of low ionic energy near track termination. Changes caused in matter by such radiation effects and their depth dependence along the ion track are important unsolved problems. Previously, depth-resolved UV- and VIS-specific luminescence $dL/dx$ for N-ions and $\alpha$-particles were measured, and the mechanisms for the formation and suppression of excimers, the effect of the helium density, and an extra peak in a Bragg-type curve were reported [1,2]. In this paper, similar measurements are done for the VUV-luminescence and the correlation with the UV- and VIS-luminescences are discussed.

2. Experimental

The track scope was composed of a 1 m imaging fiber-bundle with a square cross section of $2 \times 2$ mm where 10000 quartz fibers were bundled (Furukawa Electronic Co. Ltd), a stainless-steel slit of cross section $2 \times 0.1$ mm for beam entrance, a cryostat (modified CF-200 of Oxford Co.), and a position-sensitive photon counter, which is essentially the same as that used in ref. [1]. The depth-resolution is considered to be at least 0.02 mm. In order to detect VUV-light, the end-surface of the fiber were masked by sodium salicylate; the masking was thin and the fiber was transparent to light.

Corrected ion energy at the front-edge of the fiber was 3.687 MeV with the helium density of 0.0316 g/cm$^3$. An interference filter of band-peak 4200 Å and a cutoff filter with the edge at 5200 Å were used for the measurements of VUV- and VIS-luminescences, respectively. The sensitivity of the full system for VUV-light was corrected using a standard D$2$-lamp or by making use of approximately depth-independent VUV-luminescence of a He target at low pressure. The density of the helium target was changed from 0.02 to 0.06 g/cm$^3$; for densities larger than 0.05 g/cm$^3$, helium was liquefied. The $dE/dx$, ionic velocity, and its energy at a given track-depth were estimated using Northcliffe's table [3].

3. Results and discussion

3.1. Track-depth resolved VUV specific luminescence

Fig. 1 shows track-depth resolved VUV specific luminescence $dL/dx$, observed at various helium densities. All curves are plotted at equal heights. Each curve has a peak at the termination and the peak shifts according to the helium density. In order to exclude the effect of helium density, $dL/dx$ was plotted as a function of density-normalized residual-range with the dimension ([length] x g/cm), as shown in fig. 2. This figure shows that the peaks still shift. This suggests the existence of mechanisms for the increase and decrease in $dL/dx$ other than those attributable to the helium density. The peak positions coincided when $dL/dx$ was plotted as a function of excitation density (or energy-deposition/unit-volume), as shown in fig. 3.
These features are the same as those obtained for UV- and VIS-luminescences [2]. Therefore, the following mechanisms can be established. In the cases of UV- and VIS-luminescence, an increasing \( dL/dx \) with increasing excitation density was concluded to result from a bimolecular reaction of the lowest triplet excimers \( \tilde{3}a \). In addition, higher energy-level excimers on the right hand side cannot be produced by any other mechanism:

\[
\tilde{3}a + \tilde{3}a \rightarrow \tilde{3}d, \ \tilde{1}D, \ \tilde{1}J, \ \tilde{1}H, \ \tilde{1}A.
\]

The state \( \tilde{3}a \) is nonradiative and has a large stationary concentration since it has a long lifetime of a few ms. Also, primary excimers with higher energy relax into \( \tilde{3}a \) nonradiatively. An increase in excitation density results in a concomitant increase of \( \tilde{3}a \) so that \( \tilde{1}A \) increases more via reaction (1). The VUV-luminescence is due to the transition \( \tilde{1}A \rightarrow \tilde{1}X \). The state \( \tilde{1}A \) can also be formed by the transitions of \( \tilde{1}B \) and \( \tilde{1}C \) which are formed by the radiative transitions of the states \( \tilde{1}D, \ \tilde{1}J, \ \text{and} \ \tilde{1}H \).

With further increase in excitation density, a decrease or quenching of \( dL/dx \) shown fig. 3 may occur due to the following three-body destructive reaction (this mechanism can be rationalized by the same analysis of the experimental results for the case of VIS-luminescence reported in ref. [2]):

\[
\tilde{3}a + \tilde{3}a + \tilde{3}a \rightarrow \text{dissociative}.
\]

On the other hand, there exists a difference from the case of VIS-\( dL/dx \). In fig. 4, visible-specific luminescence \( dL/dx \) is plotted vs excitation density. Compared with the peaks in fig. 4, the peaks in fig. 3 shift to smaller excitation density. This shift is considered to

---

**Fig. 1.** VUV-specific luminescence \( dL/dx \) in helium vs the depth of the N-ion track and its density dependence. All curves are illustrated with equal maximum heights; the ordinate is expressed in an arbitrary unit. Helium densities for the bottom to the top curves are 0.062, 0.061, 0.031, 0.026, and 0.020 g/cm\(^3\), respectively, in upward direction.

**Fig. 2.** VUV-\( dL/dx \) vs N-ion range at various helium densities. This figure was obtained by converting the track-lengths of fig. 1 into the ranges using Northcliffe's table [3]. Helium densities are the same as those in fig. 1.

**Fig. 3.** VUV-\( dL/dx \) vs excitation density induced by N-ions in helium and its density dependence. The relative excitation density was estimated in the same way as in ref. [1]. Helium densities are the same as in fig. 1.

**Fig. 4.** VIS-\( dL/dx \) vs excitation density induced by N-ions in helium and its density dependence. Helium densities for the bottom curve to the top curves are 0.082, 0.080, 0.079, 0.066, 0.027, 0.024, and 0.020 g/cm\(^3\), respectively, in upward direction.
be caused by a smaller increasing of $dL/dx$ with increasing excitation density in case of VUV-luminescence compared to VIS-luminescence, because the decreasing rate in VUV-$dL/dx$ looks milder than that in VIS-$dL/dx$. This is understandable if $^1\text{C}$ and $^1\text{B}$, having long lifetimes, react with excimers such as $^3\text{a}$ before they relax into $^1\text{A}$.

3.2. Depth-resolved luminescence efficiency: VUV-$dL/dE$

As in the cases of UV- and VIS-$dL/dE$, the curves of VUV-$dL/dE$ vs excitation-density showed an additional peak, which shifted towards deeper track-depths with increasing helium density, while the peak position was independent of helium density when $dL/dE$ was plotted as a function of ionic velocity. The ionic velocity at the peak is about $4 \times 10^6$ cm/s, which agrees with that of UV- and VIS-$dL/dE$; its velocity is slightly larger than that of 1S orbital electrons in the helium atom. The $\alpha$-irradiation yielded a negligibly small peak, which is also the same for UV- and VIS-$dL/dE$. Therefore, the same mechanisms proposed in ref. [2] is applicable to VUV-$dL/dE$. One of those is the charge exchange effect: the Fano–Lichten type quasi-molecule may easily be formed since ionic velocity is slightly larger than that of orbital electrons [4]. The other is the direct excitation process of helium atoms apart from indirect processes such as ionic recombination, dimer-ion formation and relaxation to $^1\text{A}$: according to Platzmann's calculation for electron irradiated helium, a yield of the lowest singlet or triplet has a peak at the electron velocity [5] which is nearly equal to that of the present N-ion irradiation. In the present experiments, VIS-luminescence was counted using a cutoff filter in order to exclude luminescence from sodium salicylate. As a result, the additional peak of VIS-$dL/dE$ was suppressed and it was small compared with that due to VUV. Namely, the additional formation mechanism is much more effective for the formation of $^1\text{A}$.

References