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Effect of ionization density on the time dependence of luminescence from liquid argon and xenon

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The time dependence of luminescence in liquid argon and xenon has been studied for electron, α-particle, and fission-fragment excitation. The lifetimes of low excited molecular states ($^1\Sigma_u^+$ and $^3\Sigma_u^+$) do not depend on the density of excited species while the intensity ratio of $^1\Sigma_u^+$ to $^3\Sigma_u^+$ is found to be larger at higher deposited energy density. The lifetimes obtained for the $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states are 7.0 ± 1.0 nsec and 1.6 ± 0.1 μsec, respectively, in liquid argon, and 4.3 ± 0.6 and 22.0 ± 2.0 nsec, respectively, in liquid xenon. The mechanism of quenching of luminescence at a high density of excited species in liquid argon and xenon is discussed.

I. INTRODUCTION

Previously, we reported[1,2] the luminescence yield in liquid argon and xenon due to electron, α-particle, and fission-fragment excitation and found that the luminescence yield is reduced significantly under fission-fragment excitation. We attributed the reduction of the luminescence yield to quenching collisions between the excited species. In order to clarify the quenching mechanism, we have measured the decay shapes of the luminescence from liquid argon and xenon using a single-photon—counting technique.

Early studies[3,4] show that the luminescences from liquid and solid argon and liquid xenon due to electron and α-particle excitation are in the vacuum-ultraviolet (vuv) region, and exhibit no significant difference from vuv spectra taken in the high-pressure gaseous phase: a broad structureless band, width of about 10 nm centered at 128 and 172 nm for argon and xenon, respectively. The origin of the luminescence has been attributed[5–7] to low excited molecular states (self-trapped excitons), namely, the singlet state $^1\Sigma_u^+$ and the triplet state $^3\Sigma_u^+$. Since both spectra are in the same broad region as mentioned above, the two components cannot easily be separated by the difference in spectrum.[8] The decay times for $^1\Sigma_u^+$ are several nsec, and those for $^3\Sigma_u^+$ are a few μsec for condensed argon and a few tens of nsec for condensed xenon.

The present result shows that decay times for the $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states do not depend on the density of excited species along the particle track, i.e., linear energy transfer (LET), although the intensity ratios of the singlet states to the triplet states remarkably depend on LET. This fact shows that the quenching collisions must occur at a very early stage after excitation, because the decay times of molecular states would otherwise be observed to be shorter under fission-fragment excitation than under electron or α-particle excitation if the collisions between the molecular states are responsible for the reduction of the luminescence yield at a high density of excited species.

In this paper, we discuss the quenching mechanism of the luminescence at a high density of excited species and the singlet to triplet ratio with respect to LET.

II. EXPERIMENTAL METHOD

A block diagram of the single-photon—counting system used in the experiment is shown in Fig. 1. A thin source of electrons (^{207}Bi), α particles (^{210}Po and ^{252}Cf), or fission fragments (^{252}Cf) was placed in a stainless-steel vessel (6.4 cm in diameter) filled with liquid argon or xenon. The luminescence created in the liquid due to the ionizing particles is converted to visible light by *p-bis*[2-(5-phenyloxazolyl)] benzene (POPOP) which has a fast fluorescence decay time of a few nsec.

The luminescence thus converted to visible light is then detected by two photomultipliers (Hamamatsu R 329): One of them collects a large portion of the luminescence and the anode pulse is taken as a start pulse for a time-to-amplitude converter (ORTEC 467). The other photomultiplier collects a much

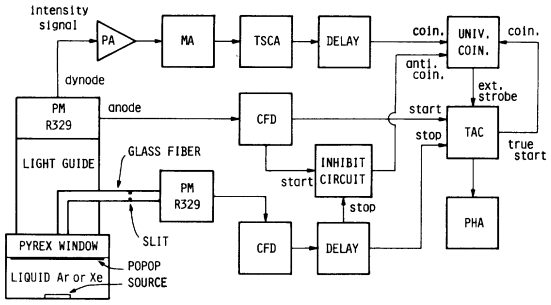

FIG. 1. Block diagram of the experimental apparatus used for the time decay measurement. PA, MA, TSCA, CFD, TAC, and PHA stand for a preamplifier, a main amplifier, a timing single-channel analyzer, a constant fraction discriminator, a time-to-amplitude converter, and a pulse-height analyzer, respectively.

smaller fraction of the luminescence through a glass fiber and a slit, and the photoelectron pulse feeds the stop input of the time-to-amplitude converter through a constant fraction discriminator and a delay unit. The discriminating level of this constant fraction discriminator was set low enough to allow single-photon detection. The stop photomultiplier was cooled by dry ice to reduce background counts.

When the probability of arrival of the stop pulse was high, corrections for photon pileup were made either by using an inhibit circuit[9,10] or mathematically.[9] The time-to-amplitude converter was carefully calibrated by a time calibrator (ORTEC 462) for all time ranges used.

In this experiment, an intensity signal is taken from the last dynode of the photomultiplier for the start pulse and pulses of energy corresponding to the energy of electrons, α particles, or fission fragments to be studied are selected by a timing single-channel analyzer and then are fed into a universal coincidence circuit and used to gate the timing pulse at the time-to-amplitude converter.

Decays were fitted to a functional form of a single exponential and a constant term, or when observed decay times were close to that of a wavelength shifter, decays were fitted to a modified form of Gale's equation,[11]

$$Y(t)=A\left\{\exp\left[\frac{\lambda^2}{4h^2}-\lambda t\right]\left[1+\operatorname{erf}\left[ht-\frac{\lambda}{2h}\right]\right]-\exp\left[\frac{\lambda_w}{4h^2}-\lambda_w t\right]\left[1+\operatorname{erf}\left[ht-\frac{\lambda_w}{2h}\right]\right]\right\}+B\,, \tag{1}$$

where λ and λ_w are the decay rates of luminescence of the liquid rare gases and a wavelength shifter, POPOP in this case, respectively, and $h=(\sqrt{2}\sigma)^{-1}$, where σ is the standard deviation of the Gaussian apparatus function. When decays include more than one exponential decay term, a linear combination of functions $Y(t)$ was used.

The instrumental resolution σ is estimated to be 0.69 ± 0.05 nsec by observing the Cerenkov radiation from uv transmitting Lucite bombarded by electrons emitted from ^{207}Bi. The fluorescence decay times $1/\lambda_w$ of POPOP were measured to be 1.43 ± 0.07 and 1.50 ± 0.07 nsec at 173 and 90 K, respectively.[12]

The vacuum system was baked out more than 74 h at a temperature above 100 °C before the experiment. The ultimate vacuum obtained was about 2×10^{-7} Torr at above 100 °C and the outgassing rate was less than 6×10^{-5} Torr/h at room temperature. The commercially available argon and xenon gases of research grade (impurity concentration is less than 10 ppm for the argon and 50 ppm including 35 ppm krypton for xenon) were further purified by using a barium-titanium getter purifier for more than a week. The argon gas was condensed by liquid oxygen (90 K) and the xenon gas was condensed by a normal hexane and cyclohexene (2:1) mixture maintained at 173 ± 2 K.

III. RESULTS

A. Liquid argon

The luminescence from liquid argon showed roughly a double exponential decay form as shown in Figs. 2 and 3. The lifetimes of the fast component, which is considered to represent the decay of the singlet state ($^1\Sigma_u^+$), are 6 ± 2, 7.1 ± 1.0, and 6.8 ± 1.0 nsec under electron, α-particle, and fission-fragment excitation, respectively, and agree within experimental errors. The lifetimes obtained for the slow component, which may be due to the triplet state ($^3\Sigma_u^+$), are 1.59 ± 0.10, 1.66 ± 0.10, and 1.55 ± 0.10 μsec under electron, α-particle, and fission-fragment excitation, respectively, and also agreed within experimental errors. The results are listed in Table I together with other experimental works.

The lifetimes observed for the $^1\Sigma_u^+$ state excited by electrons and α particles agreed with those obtained by Kubota *et al.*[13,14] but Carvalho and Klein[15] and Keto *et al.*[16] reported smaller values. The value obtained for the $^3\Sigma_u^+$ state due to electrons agreed with the value reported by Carvalho and Klein.[15] However, the values obtained by Kubota *et al.*,[13] Suemoto and Kanzaki,[17] and Keto

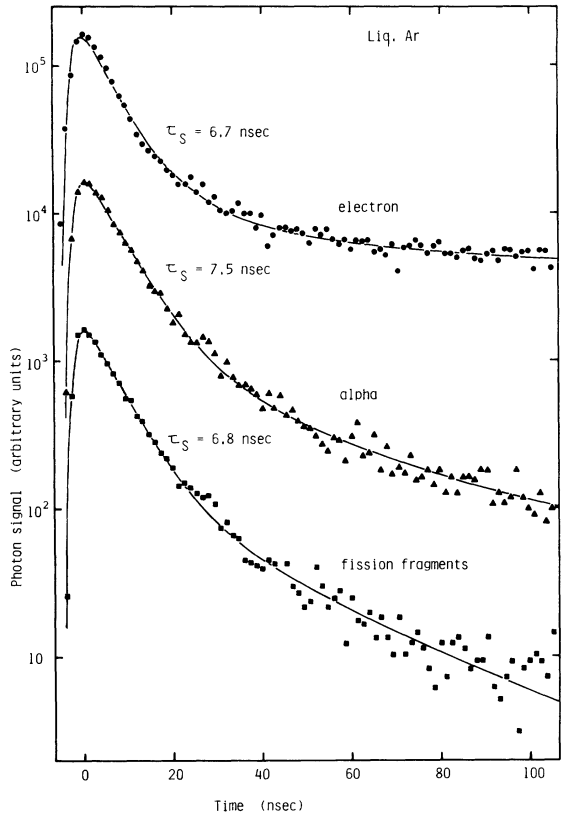

FIG. 2. Typical decay curves obtained for the luminescence from liquid argon excited by electrons (●), α particles (▲), and fission fragments (■) shown for the short time range. A slightly slow rise for electron excitation may be due to the recombination.

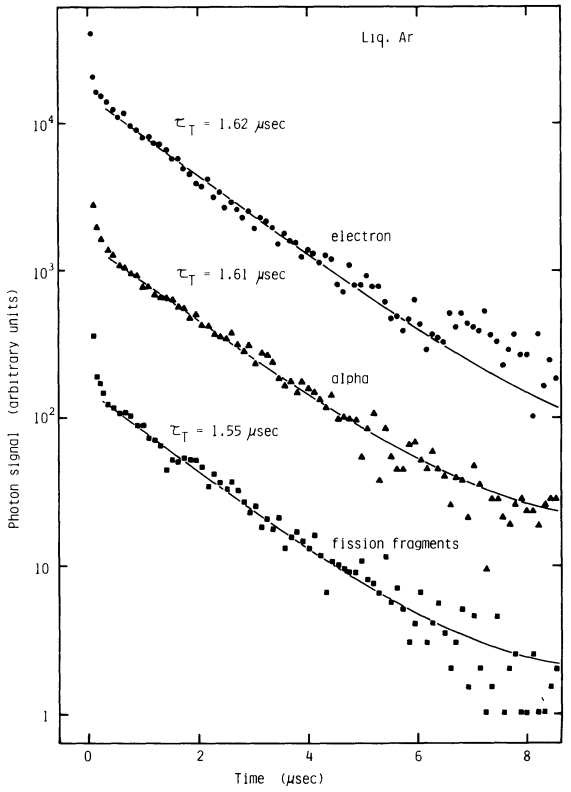

FIG. 3. Typical decay curves obtained for the luminescence from liquid argon excited by electrons (●), α particles (▲), and fission fragments (■) shown for the long time range. The fast exponential decay component is not shown in the figure.

TABLE I. Decay times for the fast τ_S and the slow τ_T components of luminescence from liquid argon. The intensity ratios I_S/I_T of the fast component to the slow component are also shown. F.F. stands for fission fragments. All decay times are in nsec.

Particle	τ_S	τ_T	I_S/I_T	Reference
Electron	6.3±0.2	1020±60	0.083	Kubota *et al.*[a]
	(5.0±0.2)	(860±30)	(0.045)	(E=6 kV/cm)[a]
	4.6	1540	0.26	Carvalho and Klein[b]
	4.18±0.2	1000±95		Keto *et al.*[c]
		1110±50		Suemoto and Kanzaki[d]
	6 ±2	1590±100	0.3	This work
α	~5	1200±100		Kubota *et al.*[e]
	4.4	1100	3.3	Carvalho and Klein[b]
	7.1±1.0	1660±100	1.3	This work
F.F.	6.8±1.0	1550±100	3	This work

[a]Reference 13.
[b]Reference 15.
[c]Reference 16.
[d]Reference 17.
[e]Reference 14.

et al.[16] are smaller than the present values. Kubota *et al.*[14] and Carvalho and Klein also reported smaller values under α-particle excitation.

The reason for this disagreement is not well understood at present. A possible reason is a difference in liquid temperature and impurities. The energy of excited states in argon is relatively large and the excited states are likely deexcited by collisions with impurities. According to our experience, nonpurified argon showed a shorter lifetime for $^3\Sigma_u^+$. Also, it has been reported that a few tens ppm of impurities such as nitrogen, oxygen, or carbon monoxide can lead to a substantial reduction in intensity of luminescence originating from low excited molecular states in condensed argon.[18]

The intensity ratios I_S/I_T of the singlet states ($^1\Sigma_u^+$) to the triplet states ($^3\Sigma_u^+$) are found to be 0.3, 1.3, and 3 for electron, α-particle, and fission-fragment excitation, respectively, and this result shows an enhancement of $^1\Sigma_u^+$ formation in higher deposited energy density. This result shows the same trend as results obtained by Kubota *et al.*[13,14] and Carvalho and Klein[15] under electron and α-particle excitation for condensed argon and xenon.

In addition to the fast (7 nsec) and the slow (1.6 μsec) components, an intermediate component which has a decay time of 20—40 nsec was observed. The intensity of this component is about 10—20% of the total intensity. This component has been reported by Kubota *et al.*[19] in the recombination luminescence under electron excitation. The origin of this component is not known at present.

B. Liquid xenon

The decay curves obtained under α-particle and fission-fragment excitation in liquid xenon showed double exponential decay forms. Typical decay curves obtained for liquid xenon are shown in Fig. 4. The lifetimes obtained for $^1\Sigma_u^+$ under α-particle and fission-fragment excitation are 4.3 ± 0.6 and 4.3 ± 0.5 nsec, respectively, and those for $^3\Sigma_u^+$ are 22 ± 1.5 and 21 ± 2 nsec, respectively, and agreed within experimental errors. The values obtained under α-particle excitation agreed with those reported by Kubota *et al.*[20] The results are listed in Table II.

The time dependence of luminescence from liquid xenon excited by electrons is shown in Figs. 4 and 5, and is quite different from those observed under α-particle and fission-fragment excitation. The curve shows a relatively slow rise and does not follow an exponential form. Kubota *et al.*[19] reported that this nonexponential component disappears when an electric field is applied. Therefore, it is clear that the nonexponential component is due to a contribution from the recombination.

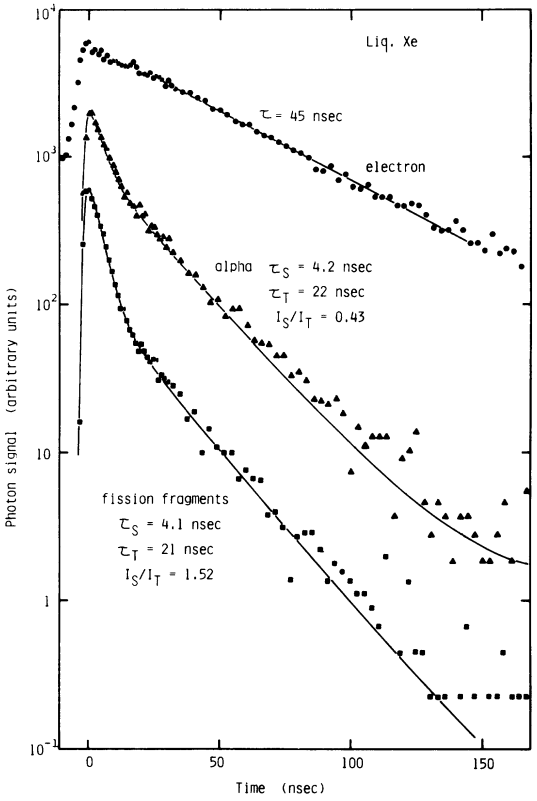

FIG. 4. Decay curves obtained for the luminescence from liquid xenon excited by electrons (●), α particles (▲), and fission fragments (■).

The ratios I_S/I_T are found to be 0.45 ± 0.7 and 1.6 ± 0.2 under α-particle and fission-fragment excitation, respectively, showing an enhancement of $^1\Sigma_u^+$ formation with higher deposited energy density.

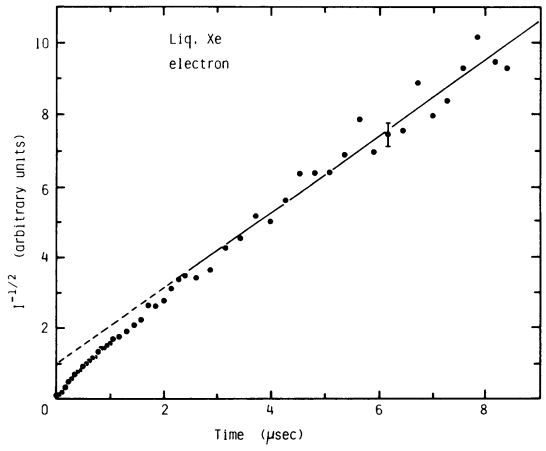

FIG. 5. Variation of $I^{-1/2}$, where I is the luminescence intensity, as a function of time obtained under electron excitation for liquid xenon. See Ref. 19.

TABLE II. Decay times for the fast τ_S and the slow τ_T components of luminescence from liquid xenon. The intensity ratios I_S/I_T of the fast component to the slow component are also shown. FF stands for fission fragments. All decay times are in nsec.

Particle	τ_S	τ_T	I_S/I_T	Reference
Electron		34±2		Kubota *et al.*[a]
	(2.2±0.3)	(27±1)	(0.05)	($E=4$ kV/cm)[a]
		32±2		Keto *et al.*[b]
		45[c]		This work
α	3	22	1.5[d]	Kubota *et al.*[e]
	4.3±0.6	22±1.5	0.45±0.07	This work
FF	4.3±0.5	21±2	1.6 ±0.2	This work

[a]Reference 13.
[b]Reference 16.
[c]Apparent decay time for times 30—150 nsec after the peak of luminescence; this component does not necessarily represent the decay for $^3\Sigma_u^+$.
[d]This value is obtained from the amplitude ratio quoted in Ref. 20.
[e]Reference 20.

IV. DISCUSSION

Ionizing particles introduced in condensed rare gases such as liquid argon and xenon produce excitons and electron-ion pairs along the track. Free excitons produced form excited molecules R_2^* through the self-trapping process within a few picoseconds.[6] The ions are localized through the formation of molecular ions R_2^+ also within a few picoseconds.[21,22] The electrons emitted lose their kinetic energy through production of excitons and electron-ion pairs, and then are thermalized through phonon interaction. Then a part of the thermalized electrons recombine with molecular ions and the remaining part escapes recombination. The excitons and the electron-ion pairs finally go to the lowest excited molecular state $^1\Sigma_u^+$ or $^3\Sigma_u^+$ and decay radiatively to the repulsive ground state $^1\Sigma_g^+$. Although the transition from $^3\Sigma_u^+$ to $^1\Sigma_g^+$ is forbidden by the selection rule $\Delta S=0$, strictly speaking, the Russell-Saunders notation is no longer an adequate representation for heavy atoms such as argon and xenon. It is due to this fact that $^3\Sigma_u^+$ decays radiatively to $^1\Sigma_g^+$.

The present result shows that the decay times of the fast and slow components in the decay of luminescence, corresponding to the decay of $^1\Sigma_u^+$ and $^3\Sigma_u^+$, respectively, do not change while the luminescence yield is reduced significantly for fission-fragment excitation[1,2] and the singlet to triplet ratios I_S/I_T increase with LET.

No information is available at present for the luminescence spectra of condensed rare gases due to ions heavier than α particles. However, we assume here that the low excited molecular states (self-trapped excitons) $^1\Sigma_u^+$ and $^3\Sigma_u^+$ are responsible for the luminescence from liquid argon and xenon under fission-fragment excitation, since the luminescence is in the vacuum-ultraviolet region[1] and the decay times observed are the same as those measured under α-particle excitation.

Few studies have been made for the luminescence from condensed rare gases due to ions heavier than α particles. Mutterer[23] has observed the time decay of the luminescence from solid xenon excited by ^{50}Ti ions of 1.4 MeV/amu which is larger than the energy per amu of fission fragments and reported that the decay exhibits two exponential decays of 4.7 and 22.2 nsec which agreed with the values measured under α-particle excitation and also with present values obtained for liquid xenon. He also reported that the decay has a longer component, delayed by $\sim$150 nsec. This longer component is neither observed under α-particle excitation[20] nor in liquid xenon under α-particle and fission-fragment excitation. This component may be attributed to δ rays produced by ^{50}Ti ions passing through solid xenon. Since Ti ions of this energy can produce δ rays of relatively high energy, the luminescence may include some electron excitation characteristics, e.g., the existence of a long decay component due to the recombination.

When condensed rare gases are excited by fission fragments very high concentrations of excited and ionic species are produced along the track. The collision between two excited species would emit an electron which has a kinetic energy close to the difference between twice the excitation energy E_{ex} and the band-gap energy E_g (i.e., $2E_{ex}-E_g$). This electron can lose energy through a rapid energy-loss

process[24] before recombination or can escape recombination. This process could be a main mechanism for the reduction of the luminescence yield.[1]

The lifetimes obtained here for the $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states under fission-fragment excitation are the same as those obtained under electron and α-particle excitation, so we rule out the collisions between those low excited molecular states ($v'=0$) from the quenching collisions. Then free excitons and/or highly excited species may be responsible for the quenching. One may be surprised that collisions between free excitons, and not self-trapped excitons, may be responsible for the quenching while excitons are self-trapped within a few picoseconds and self-trapped excitons have lifetimes of several nanoseconds to a few microseconds. However, this idea may be supported by the fact that the lifetimes of free excitons are long enough to cause Penning ionization in liquid argon-xenon mixtures.[25]

The luminescence yields per unit energy deposit due to fission fragments are $\frac{1}{4}$ and $\frac{1}{3}$ those due to α particles for liquid argon and xenon, respectively.[1,2] The ratios I_S/I_T due to α particles and fission fragments are 1.3 and 3, respectively, for liquid argon, and those for liquid xenon are 0.45 ± 0.07 and 1.6 ± 0.2, respectively. From these facts, it is plausible to think that both the singlet and the triplet are quenched.

The mechanisms which may be responsible for the increase of the ratio I_S/I_T at a high LET are the following: (1) preferential excitation of the singlet state at a high LET, (2) relaxation of spin in the recombination process, (3) spur recombination, (4) triplet-triplet annihilation, (5) collision of two excited triplet states which results in two excited singlet states,[15] and (6) superelastic collisions with thermal electrons.

Process (1) does not seem to explain the result. The light and fast particles likely excite more singlet states than the slow and heavy particles do. If this is the main mechanism which controls the I_S/I_T ratio, the result should be just the opposite. Process (2) would give the same trend as the experimental one. Relaxation of spin in this process is induced by magnetic interactions. No information is available for the spin-relaxation time in liquid rare gases, but it would be long compared with the recombination time ($\lesssim 1$ nsec for liquid argon).

I_S/I_T decreases with LET in organic liquids[26] and this has been partly explained by spur-recombination processes.[27] Since the recombination time is much shorter than the spin-relaxation time in many organic liquids, spin conservation applies to the recombination process. In low LET, electrons and ions recombine through geminate recombination, which favors the production of the singlet state. In high LET, homogeneous recombination becomes the main process. Electrons and ions recombine at random, then I_S/I_T will be roughly $\frac{1}{3}$ (statistical weight). However, this explanation apparently cannot be applied to liquid rare gases.

Under certain conditions, two long-lived triplet states may collide with one another in organic solution and "annihilate" each other simultaneously producing one in the excited singlet state and the other in a ground state.[28] This process (4) is not applied to rare gases. Collision of this type between two excited states, both free excitons and molecular states, will result in ionization in rare gases ($2E_{ex} > E_g$) and such collisions occur in times much shorter than the radiative lifetimes as observed here. Therefore, one might expect that the singlet-singlet quenching happens as well. Process (5) may be excluded too, since the reverse process may be expected with a similar probability.

It is well known that in the gaseous state the singlet state is changed to the triplet state by superelastic collisions with thermal electrons.[29] The recombination process is very fast when condensed rare gases are excited by α particles and fission fragments; however, in electron excitation, recombination occurs relatively slowly. Then these electrons could quench the singlet states to the triplet states by collisions. This process (6) is most plausible to explain the LET dependence of the singlet-to-triplet ratio I_S/I_T in liquid rare gases although process (2) is also possible.

Itoh[30] has studied the LET effect on the luminescence from alkali halide crystals such as NaCl, KBr, and KI using 2-MeV H^+ and He^+ ions and observed the decrease of luminescence yield with LET as in liquid argon and xenon. Kimura *et al.*[31] measured the decay and time-resolved spectra of the emission from KBr due to heavy ions. They reported that the lifetimes for $\sigma-(^1\Sigma_u)$ and $\pi-(^3\Sigma_u)$ excitons (self-trapped excitons) are similar to those reported for photon,[32] electron,[33] and proton[34] excitation while the intensity ratio of σ emission to π emission (I_S/I_T) is enhanced by a factor as large as 5. This LET effect observed in ionic crystals follows the same trend as that observed in liquid argon and xenon.

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