Specific-ionization–density effect on the time dependence of luminescence in liquid xenon

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We have investigated the influence of specific ionization density on the time dependence of luminescence in liquid xenon. The experiments have revealed that the luminescence decay is governed by the decay rates of two excited molecular states ($^1\Sigma_u^+$ and $^3\Sigma_u^+$) for alpha-particle excitation (high specific-ionization density) and by the recombination rate of free electrons and molecular ions for high-energy electron excitation (low specific-ionization density). A significant enhancement of the formation of $^1\Sigma_u^+$ states was observed for alpha-particle excitation.

In a recent article[1] (hereafter referred to as I), we reported the investigation of the time dependence of the luminescence due to the recombination process of electrons and localized molecular ions R_2^+ in liquid argon, krypton, and xenon excited by high energy electrons. The two lowest excited molecular states ($^1\Sigma_u^+$ and $^3\Sigma_u^+$) responsible for the luminescence are formed through a recombination process. The time dependence of the recombination luminescence depends on the kinetic characteristics of the recombination process of thermalized electrons and R_2^+ and the lifetimes τ_1 and τ_2 of the two excited molecular states, $^1\Sigma_u^+$ and $^3\Sigma_u^+$, respectively. The lifetimes τ_1 and τ_2 for luminescence from liquid argon, krypton, and xenon excited by high-energy electrons have been studied with such a high field that all of the observed decay characteristics have to be attributed to the self-trapped exciton luminescence in Ref. 2.

It was found in I that the recombination luminescence decay for liquid argon showed two exponential-decay components whose decay times were nearly equal to the lifetimes τ_1 and τ_2, and it was concluded that the characteristic recombination time T_r ($=1/n_0\alpha$) is much shorter than τ_1 and τ_2. Here n_0 and α are the initial number density of electron-hole pairs and the coefficient of the e-R_2^+ recombination. On the other hand, for liquid xenon, the recombination luminescence decay is governed by T_r which is longer than τ_1. This fact suggested that the recombination cross section for liquid xenon is about 700 times smaller than that for liquid argon.

The initial rise of the recombination luminescence was too fast to be followed experimentally in I. However, by using Eq. (5) in I, it was shown analytically that the initial rise is governed by T_r for $T_r < 2\tau_1$ (for liquid argon) and τ_1 for $T_r > 2\tau_1$ (for liquid xenon).[3]

Here we consider the luminescence decay of liquid xenon excited by alpha particles. Because of its high specific ionization (about 1000 times) and excitation density for alpha-particle excitation, compared with that for high-energy electrons, it is expected that T_r is much shorter than τ_1 and τ_2 and hence the luminescence decay is not governed by T_r, but by τ_1 and τ_2. Thus luminescence of liquid xenon excited with high specific-ionization density should have two exponential decays whose lifetimes are τ_1 and τ_2. The present experiment shows that this is the case.

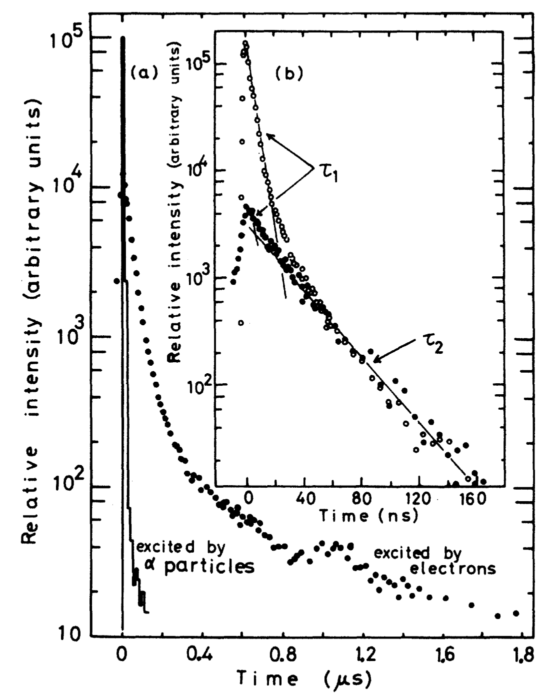

FIG. 1. Time dependence of the luminescence intensity for liquid xenon excited by alpha particles and by electrons for the long time range (a). Time dependence for alpha-particle excitation (○) and that of the self-trapped exciton luminescence excited by electrons (●) are indicated for the short time range (b).

The experimental details have been reported in I. Instead of a ^{207}Bi internal-conversion electron source, a ^{210}Po alpha source was used. Figure 1(a) shows the decay curves of liquid xenon excited by alpha particles and by high-energy electrons. A significant difference in decay curves was observed for different specific-ionization density. Figure 1(b) shows the decay curve obtained by alpha-particle excitation for the short time range and the decay curve of self-trapped exciton luminescence excited by high-energy electrons. The latter was observed by applying an electric field of 3 kV/cm. It is clearly seen that the decay excited by alpha particles exhibits two decay components, whose decay times are 4 and 27 ns. These decay times are nearly equal to the τ_1 and τ_2 values for self-trapped exciton luminescence excited by electrons.[2] Since the luminescence decay for alpha-particle excitation is not governed by T_r but by τ_1 and τ_2, it is concluded that T_r is much shorter than τ_1, as we expected. Figures 1(a) and 1(b) clearly show that the relative intensity of the fast component increases with increasing specific-ionization density.

The ratio $(A_1/A_2)_\alpha$ of photon numbers from $^1\Sigma_u^+$ state to that from the $^3\Sigma_u^+$ state of liquid xenon excited by alpha particles is estimated to be about 7.5 from Fig. 1(b), which is about ten times higher than the value of 0.8 ± 0.2, corresponding to electron excitation from Ref. 1.

The possible mechanisms that may be responsible for the higher value of $(A_1/A_2)_\alpha$ are (1) a large probability of radiationless annihilation of the $^3\Sigma_u^+$ states which are formed at high density by alpha-particle excitation and (2) preferential formation of the $^1\Sigma_u^+$ states.

If the high value of the ratio is due to process (1), we must have a short lifetime for $^3\Sigma_u^+$ states and also have a decrease of the luminescence intensity for alpha-particle excitation compared with electron excitation. Also, as shown in Fig. 1(b), the slow lifetime for alpha excitation is the same as that for electron excitation within experimental error.

In an auxiliary experiment, pulse-height distributions were measured for a mixed ^{210}Po and ^{207}Bi source under the condition that the photomultiplier current was integrated over a period of 10 μs. From the pulse-height data we find that the ratio L_α/L_β is 6.1, where L_α and L_β are the pulse heights for 5.3-MeV α particles ($=E_\alpha$) and for electrons of energy (E_β) about 1 MeV, which are composed of 1.06-MeV L-conversion electrons and 0.976-MeV L-conversion electrons. This value implies that the intensities per unit energy are in the ratio $(L_\alpha/E_\alpha)(L_\beta/E_\beta)^{-1} = 1.15$, and hence the luminescence intensity is proportional to the particle energy dissipated in the liquid xenon. These experimental results lead to the conclusion that process (1) described above should be ruled out and that process (2) is plausible.

It was shown in I that 74% of the luminescence intensity of liquid xenon is due to the emission of $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states formed from the dissociative recombination process $R_2^+ + e \rightleftarrows R_2^{**} + R$ and $R^{**} + R \rightarrow R_2^*$. In alpha-particle excitation highly excited neutral molecules R_2^{**} and excited exciton states R^{**} are formed densely in the vicinity of the track and collide with each other, and hence $^1\Sigma_u^+$ and $^3\Sigma_u^+$ states should be formed in a ratio that differs from the case of electron excitation.

The study of the enhancement of $^1\Sigma_u^+$-state formation by heavy-ion excitation in the liquid rare gases is interesting, both theoretically and experimentally. The enhancement of $^1\Sigma_u^+$-state formation under alpha-particle excitation has also been reported for liquid argon by Kubota *et al.*[4] and Carvalho and Klein,[5] and, for KBr by Kimura and Imamura.[6]

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