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Recombination luminescence in liquid argon and in liquid xenon

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An electric-field effect on the luminescence of liquid argon and xenon excited by ^{207}Bi internal-conversion electrons has been studied. A decrease in the luminescence was observed (67% at 10 kV/cm for liquid argon and 74% at 12.7 kV/cm for liquid xenon relative to the intensity without an applied field), and is explained by the escape of electrons and holes before recombination. This provides strong evidence for the existence of the recombination luminescence. Quantum efficiencies of recombination and self-trapped exciton luminescence are briefly considered.

I. INTRODUCTION

Vacuum-ultraviolet luminescence from solid and liquid argon, krypton, and xenon has previously been studied extensively for uv light, x rays, and ionizing charged-particle excitation.[1-6] In all of these investigations, a narrow-band intrinsic luminescence was observed, which was explained as the radiative decay of an excited molecule. Although Martin[7] and Molchanov[8] suggested that this molecule can be produced either by self-trapping of a free exciton or by recombination of a self-trapped hole and a free electron, quantitative information has been lacking.

In order to confirm the role of recombination in the luminescence, we applied an electric field and measured the luminescence intensity of liquid argon and liquid xenon excited by internal-conversion electrons from ^{207}Bi. Simultaneously, free electrons were collected by a pulse technique in order to obtain the unrecombined charge.[9,10] The present experiment shows that a large fraction of the luminescence comes from a recombination process. In addition, information regarding the nonradiative process in the bulk is obtained.

II. EXPERIMENTAL PROCEDURE

To observe the electronic-field effect on the luminescence, a parallel-plate ionization chamber with a glass window was used. The cathode is a flat-surfaced disk with the diameter of 27.5 mm. The collector is an array of wires with a diameter of 10 μm strung with a 100-μm spacing onto a circular flange. The distances between the cathode and the collector are 5.0 and 3.2 mm for liquid argon and liquid xenon, respectively. A ^{207}Bi conversion electron source (2-mm-diam spot), which emits electrons with energies of 1.05, 0.976, 0.55, and 0.48 MeV, was deposited chemically on the center of the cathode. The chamber was baked at about 120 °C for more than 48 h and the ultimate vacuum obtained was 3×10^{-7} Torr, with an outgassing rate less than 2×10^{-8} Torr l/sec.

To observe the ultraviolet luminescence, the inner surface of the window was coated with sodium salicylate. It was used because of its uniform response over a broad wavelength range (500–2500 Å).[11] The output pulse from the photomultiplier (RCA 8575) was clipped through a resistance-capacity (RC) network with a clipping time of 5 μsec, and then sent through a pulse shaper into a pulse-height analyzer. For measurements of ionization, signal pulses from the collector were amplified by a charge-sensitive amplifier and a main amplifier with differential and integrating time constants of 2 μsec.

High-purity xenon gas (obtained from Toshiba Co. Ltd.) was purified with barium-titanium getters maintained at about 600 °C; details of the purifier have been published previously.[12] The purified xenon was condensed into the chamber which was

cooled by an n-propyl alcohol bath maintained at 183 K, corresponding to a xenon pressure of 2.5 atm. High-purity argon gas (obtained from Toshiba Co. Ltd. and containing H_2, 2 ppm; O_2, 0.9 ppm; C_nH_m, 0.2 ppm, and undetected amounts of N_2 and CO_2) was liquified into the chamber cooled with liquid oxygen refrigerant to 99 K, corresponding to an argon pressure of 3 atm.

III. EXPERIMENTAL RESULTS

Figure 1 shows typical pulse-height spectra for the luminescence of liquid argon excited by ^{207}Bi conversion electrons with and without an applied electric field. The peak in each spectrum is due to the complex of K- and L-conversion electrons of energies 0.976 and 1.05 MeV, respectively. The reproducible results were obtained for different experimental runs. For all cases in this experiment, the electric field produced a decrease in luminescence intensity.

Figure 2 shows typical pulse-height spectra for the ionization of liquid argon excited by ^{207}Bi conversion electrons. As seen in the figure, the peak due to the complex of K- and L-conversion

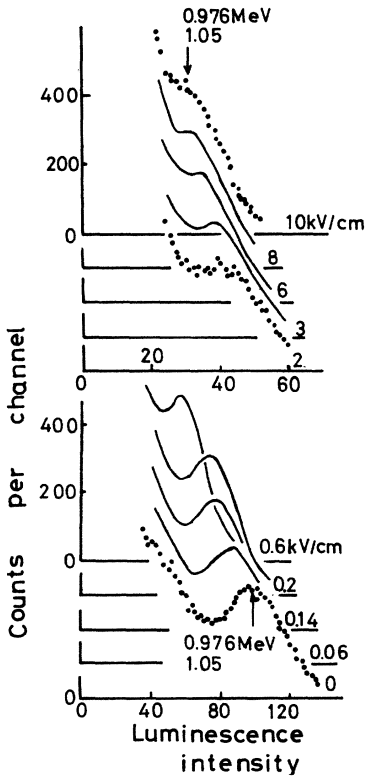

FIG. 1. Luminescence pulse-height distributions in liquid argon excited by ^{207}Bi conversion electrons for various electric-field strengths. Note the change in scale of the abscissa.

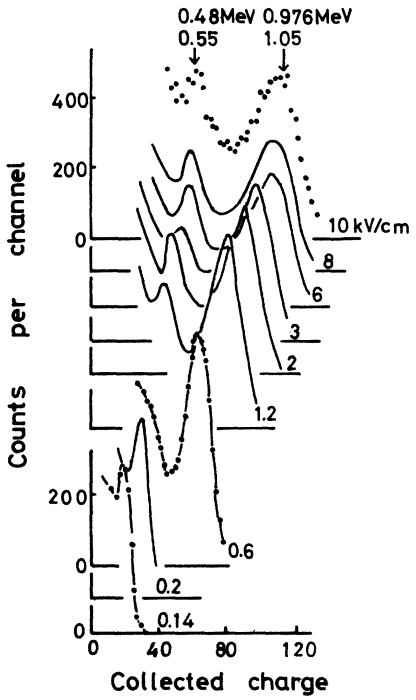

FIG. 2. Ionization pulse-height distributions in liquid argon excited by ^{207}Bi conversion electrons for various electric-field strengths.

electrons of energies 0.48 and 0.55 MeV is also observed for the electric field strength of higher than 1.2 kV/cm.

Figure 3 shows the measured peak pulse heights of the luminescence L (in the case of liquid xenon not only conversion electrons but also photoelectric electrons are included) versus field strength $\mathcal{E}$ together with the measured peak pulse heights Q of the ionization for the complex of 0.976- and 1.05-MeV electrons for liquid argon and liquid xenon. The error in the measurements at each

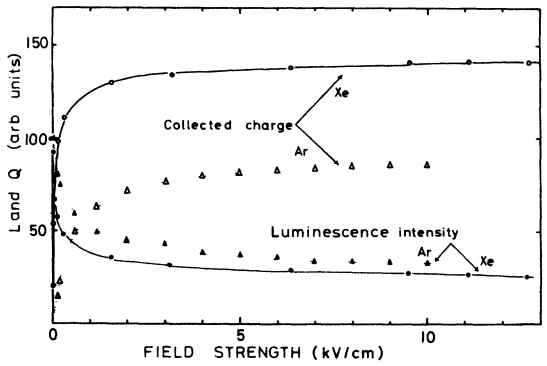

FIG. 3. Variations of relative luminescence intensity L and collected charge Q in liquid argon and in liquid xenon against applied electric-field strength for 0.976- and 1.05-MeV electrons.

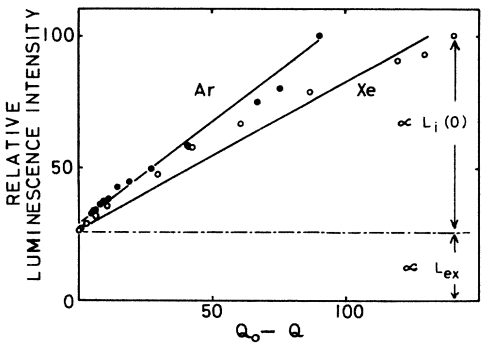

FIG. 4. Relative luminescence intensity against uncollected charge Q_0-Q.

point in Fig. 3 is estimated to be about ±2%. As seen in Fig. 3, the observed luminescence for liquid xenon decreases with the field and then approaches a saturation value. A similar trend was also found for liquid argon. The decreases in the luminescence were (74 ± 2)% at 12.7 kV/cm for liquid xenon and (67 ± 2)% at 10.0 kV/cm for liquid argon. Also shown in the figure are saturation characteristics for collected charge in liquid xenon and liquid argon. At the highest field, 99% and 95% of the produced charge for liquid xenon and liquid argon, respectively, were collected.

The intensity of recombination luminescence can be regarded as proportional to the uncollected charge $Q_0 - Q$, where Q_0 is the pulse height for the produced total charge. Figure 4 shows the variation of the luminescence intensity L against $Q_0 - Q$ as obtained from Fig. 3. Here, we have adopted the value of Q at 12.7 kV/cm as Q_0 for liquid xenon. For liquid argon, Q_0 was estimated from plots of $\mathcal{E}^{-1}$ against Q^{-1}, which has been used often in the analysis of the liquid-ionization chamber,[13,14] and the plotted points in Fig. 4 lie reasonably well on straight lines. This evidently shows that the decrease in luminescence with an applied field is due to the escape of electron and holes from recombination.

IV. DISCUSSION

The simultaneous decrease in luminescence intensity and the increase in collected electrons indicate that a large fraction of luminescence intensity is produced from a recombination process. We consider that the field-dependent intensity $L_i(\mathcal{E})$ is due to the recombination luminescence and the field-independent intensity L_{ex} to the luminescence from self-trapped excitons. The ratio $L_{ex}/L_i(0)$ can be obtained from Fig. 4 and this is shown in the first column of Table I.

The ratio N_{ex}/N_i of the number of excitons to electron-hole pairs produced by ionizing charged

TABLE I. $L_{ex}/L_i(0)$, N_{ex}/N_i, and η_{ex}/η_i for liquid argon and for liquid xenon.

Liquid	$\frac{L_{ex}}{L_i(0)}$	N_{ex}/N_i calculated	N_{ex}/N_i measured	$\frac{\eta_{ex}}{\eta_i}$
Ar	0.43	0.21	0.19 ± 0.02 [a]	2
Xe	0.33	0.06	···	5

[a] Reference 15.

particles has been calculated as shown in the second column of Table I; details of the calculation have been published previously.[9,10] This calculation is made based on the following assumptions:

(i) The probability of producing an excited state of excitation energy E by ionizing particles is proportional to $f(E)/E$, where $f(E)$ is the oscillator strength.

(ii) The electronic band structure of the liquid is almost the same as that of the solid. The validity of these assumptions has been experimentally supported by the observation of exciton-enhanced ionization from Xe dopant in liquid argon.[15]

The quantum efficiencies η_{ex} and η_i of exciton and recombination luminescence are defined by the relations $L_{ex} = \eta_{ex} N_{ex}$ and $L_i(0) = \eta_i N_i$. Ratios of η_{ex}/η_i have been obtained from $L_{ex} N_i / L_i(0) N_{ex}$, by using the measured $L_{ex}/L_i(0)$ values and the calculated values of N_{ex}/N_i. The results are shown in the last column of Table I and they show that η_i is significantly smaller than η_{ex}.

Brodman *et al.*[6] have reported a low η_i for solid xenon excited by photons of energies higher than the band-gap energy of solid xenon. This was explained by the process of photoemission from the surface competing with recombination luminescence. In this experiment we have found a large value of η_{ex}/η_i even in the bulk. Here it should be pointed out that the photoemission is not only a competitive process with recombination luminescence, but that other nonradiative processes may play an important role. The problem of nonradiative processes in the bulk is interesting, both theoretical and experimental.

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