

Mechanism of Scintillation of Helium, Helium-Argon, and Helium-Neon Gas Mixtures Excited by Alpha Particles

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An experimental investigation of the mechanism of scintillation of helium, helium-argon, and helium-neon mixtures excited by α particles has been performed. No detectable decrease in light yield was observed at pressures less than 3 atm when the applied electric field was increased in steps to $E/p \sim 1.0$ V/cm Torr, where E is the electric field and p is the gas pressure. The pulse shape of helium scintillation light consists of a slow component and a spike appearing on the leading edge of the pulse. The main component of light intensity is represented by the formula $\exp(-t/\tau_m) - \exp(-t/\tau_f)$, where t is the time in sec, $\tau_m = 10 \times 10^{-6}$ sec, and $\tau_f = 0.3p^{-2.2 \pm 0.3}$ sec (p in Torr). Most of the emitted photons had a wavelength of less than 1050 Å. On the basis of these results, it is concluded that the emitted photons are released in the decay of excited helium molecules formed as a result of a three-body collision between a metastable and two ground-state helium atoms. A characteristic large drop in the light yield for a mixture of a small proportion of argon in a major fraction of helium can be explained by the production of argon ions by metastable helium atoms (Penning process). The same characteristic large drop for a mixture of small concentration of neon in helium seems to be caused by the excitation transfer from helium to neon atoms. The cross section σ_f for the formation of an excited helium molecule by a three-body collision is estimated to be $50 \times 10^{-22} p$ cm² at 300°K, where p is in Torr. From the value of σ_f , the cross section σ_i of the helium-argon Penning process is calculated as 41×10^{-16} and 14×10^{-16} cm² by using the ratios of σ_i to σ_f estimated by Jesse and Sadauskis and by Northrop and Gursky, respectively.

I. INTRODUCTION

THE inert gases have been often used as scintillators for the detection of heavy charged particles. The mechanism of the inert-gas scintillation, however, is not yet well understood because of the complexity of the scintillation process.

In 1958, Northrop and Gursky¹ measured the relative scintillation yield of binary mixtures of inert gases due to α particles using the wavelength shifters diphenyl stilbene and quaterphenyl; they found that the light yield showed a characteristic large drop for a mixture containing a small proportion of heavier gas in a major fraction of the lighter. To interpret this effect, they proposed that the atomic ions produced by α particles collided with normal atoms, resulting in stable molecular ions which subsequently recombined with electrons emitting photons as scintillation light. Recently, Esterling and Lipman,² who measured the pressure dependence of the light yield of helium-gas scintillation in a pressure range of 2 to 40 atm, deduced that the scintillation light was emitted in the recombination process.

On the other hand, Ward³ and Bennett⁴ have demonstrated experimentally that the scintillation light due to

recombination is negligibly small in the visible wavelength region for helium excited by α particles at ordinary pressures (0.5~1 atm). Tavendale⁵ has studied the effect of an electric field on the scintillation of an argon-nitrogen gas mixture at 2 atm, and showed that the light yield from recombination was only 3% of the total for α -particle excitation. From these experimental facts, it may be expected that the contribution of the recombination process to the scintillation of an inert gas excited by α particles would be very small at ordinary pressures. This is inconsistent with the interpretation of Northrop and Gursky¹ and of Esterling and Lipman.² In the experiments of Ward,³ of Bennett,⁴ and of Tavendale,⁵ however, the ultraviolet light, the main component of inert-gas scintillation, could scarcely have been observed, because an efficient wavelength shifter for the ultraviolet light was not used.

In order to clarify the mechanism of inert-gas scintillation in the ultraviolet region, we attempted to measure the electric field effect on the light yield, the pressure dependence of the pulse shape of helium scintillation light, and the relative light for helium-argon and helium-neon gas mixtures using wavelength shifters for the ultraviolet light. The mechanism of inert-gas scintillation is discussed in this paper.

¹ J. A. Northrop and J. C. Gursky, Nucl. Instr. Methods **3**, 207 (1958).

² R. J. Esterling and N. H. Lipman, Rev. Sci. Instr. **36**, 493 (1965).

³ A. Ward, Proc. Phys. Soc. (London) **A67**, 841 (1954).

⁴ W. R. Bennett, Jr., Ann. Phys. (N. Y.) **18**, 367 (1962).

⁵ A. F. Tavendale, Proc. Phys. Soc. (London) **79**, 652 (1962).

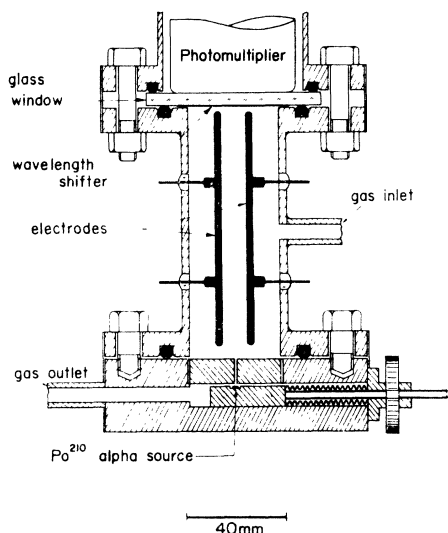


FIG. 1. Sectional view of the vessel used for the measurement of gas scintillation.

II. EXPERIMENTAL PROCEDURE

A. Scintillation Vessel

A cylindrical stainless-steel vessel, 36 mm i.d. and 110 mm in depth, was used in this experiment as shown in Fig. 1. The inner surface of the vessel was coated with magnesium oxide for the purpose of reflecting as much light as possible. The effect of the outgas from the magnesium oxide on the purity of the gas was estimated to be less than 100 ppm, as will be described in Sec. II C. A glass plate was mounted at the upper end of the vessel as a window for a photomultiplier. Two stainless-steel rectangular electrodes (24 mm \times 92 mm) were fixed 10 mm apart to the wall of the vessel parallel with its axis. A Po^{210} α source could be inserted and removed from the center of the lower end of the vessel without breaking the vacuum. The α particles were collimated along the axial line of the vessel in order to prevent them from reaching the electrodes. When the vessel was used at pressures greater than 1.87 atm of helium, α particles did not reach the window of the vessel.

B. Wavelength Shifters

To observe the ultraviolet light, the inner surface of the window was coated with a wavelength shifter. Two kinds of wavelength shifters, diphenyl stilbene and sodium salicylate, were used. Although the spectral response of the emission of diphenyl stilbene induced by vacuum ultraviolet photons and its fluorescence decay time were not accurately known, it was used for the measurement of the light yield of scintillation of the gas mixture to ensure the same experimental conditions as those of Northrop and Gursky, who used diphenyl stilbene as a wavelength shifter. Diphenyl stilbene was slowly evaporated onto the window of the vessel in a

vacuum of less than 2×10^{-5} Torr. The optimum thickness for light yield was found to be about $250 \mu\text{g}/\text{cm}^2$.

For the observation of the pulse shape of the scintillation light, sodium salicylate was employed because of its uniform response over a broad wavelength region (500 \sim 2500 Å)⁶ and its fast fluorescence decay time ($8.5 \sim 10.0 \times 10^{-9}$ sec),⁷ which is short enough to observe the pulse shape of helium scintillation light. The window was coated with sodium salicylate dissolved in methyl alcohol, brushed on to a thickness of about $1 \text{ mg}/\text{cm}^2$, which is the optimum for a fluorescence converter according to Seya and Masuda.⁸

C. Gas-Filling System

In Fig. 2 is shown the gas-filling system, made of glass, and capable of withstanding 3 atm pressure. Viton O-ring gaskets and Teflon packings were used. The gas pressure was measured by a Bourdon tube calibrated with a U-tube mercury manometer. Before filling, the charcoal traps in the system were baked out at $400 \sim 450^\circ\text{C}$ for more than 12 h, and the system and the vessel were evacuated to 2×10^{-6} Torr or less. Reagent-grade helium was slowly fed into the vessel through two charcoal traps in series cooled by liquid nitrogen. A glass circulating pump similar to that of Bennett⁹ was used to maintain the high purity of the gas. When the circulation was stopped, the ion current produced by α particles gradually increased to about 1.3 times the initial value. This fact seems to show that the concentration of the impurity mainly from wavelength shifter and light reflector was less than 100 ppm during circulation.¹⁰

D. Electronics

The photomultiplier used was of the type RCA-6810A. The output pulse from the photomultiplier was clipped through the RC network, which consisted of a

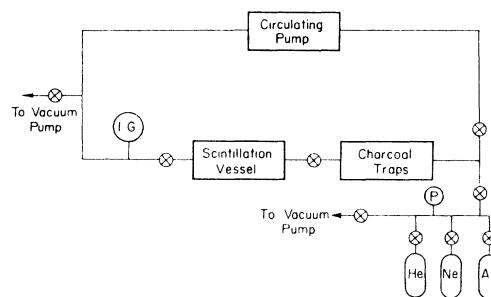


FIG. 2. Schematic diagram of gas filling system. I.G., ionization gauge; P, pressure gauge (Bourdon tube).

⁶ A. Becker, Max-Planck Institute for Physics and Astrophysics Report No. MPA-18/63 (1963) (unpublished).

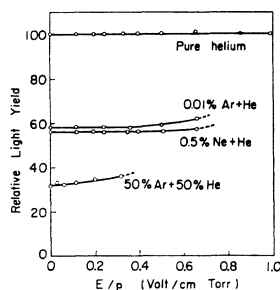
⁷ G. K. Herb and W. J. Van Sciver, *Rev. Sci. Instr.* **36**, 1650 (1965).

⁸ M. Seya and F. Masuda, *Sci. Light (Japan)*, **12**, 9 (1963).

⁹ W. R. Bennett, Jr. *Rev. Sci. Instr.* **28**, 1092 (1957).

¹⁰ W. P. Jesse and J. Sadauskis, *Phys. Rev.* **100**, 1755 (1955).

FIG. 3. The electric field effect on the slow component of scintillation light yield. Clipping time: 10×10^{-5} sec.



photomultiplier load resistance and a stray capacitance. For the measurement of the light yield of the slow component of the scintillation, clipping times of 0.5×10^{-6} and 10×10^{-5} sec were used. For the measurement of the fast component of scintillation, the output current pulse was fed through a distributed amplifier and was clipped with an RC network of 35×10^{-9} sec. These output pulses were sent through a pulse shaper into a 400-channel pulse-height analyzer. For the sensitive detection of electric-field effect, the output pulses were also fed through a single-channel pulse-height analyzer by setting the integral discriminator at a point where about $\frac{1}{2}$ of the total particle rate was registered. This method could easily detect a change in pulse height of more than 1%. The reason for the use of 0.5×10^{-6} sec as the clipping time was to establish the same experimental conditions as those of Northrop and Gursky. The slow clipping time, 10×10^{-5} sec, was used to detect the long tail of the pulse of helium, helium-argon, and helium-neon scintillation, which extends to the order of 10^{-5} sec.

For observation of the pulse shape, a clipping time of 5×10^{-9} sec was used. The output current pulse was fed through a cathode follower (rise time 4.5×10^{-9} sec) into a distributed amplifier with a bandwidth of 200 Mc/sec, and was observed by a fast oscilloscope with a bandwidth of 100 Mc/sec. The total rise time of the electronic system was estimated to be about 6×10^{-9} sec. The ion current produced by α particles was measured by the use of a vibrating-reed electrometer.

III. EXPERIMENTAL RESULTS

A. Pure Helium

When the window was not coated with a wavelength shifter, no helium scintillation was observed at pressures less than 3.0 atm. When the inner surface of the window was coated with diphenyl stilbene or sodium salicylate, the pulse height increased and the spectrum became a peak when the pressure was higher than about 0.7 atm. The full width at half-maximum (FWHM) of the peak was 30% at 2 atm.

The electric-field effect on the slow component of the scintillation light was investigated by using clipping times of 0.5×10^{-6} and 10×10^{-5} sec. The effect on the fast component, occurring within the time 50×10^{-9} sec,

was also studied by clipping the output current pulse of a distributed amplifier with an RC network of 35×10^{-9} sec. A potential difference was applied up to 1500 V between the electrodes and the maximum value of E/p was about 1.0 V/cm Torr, where E is the electric field and p is the gas pressure. The ion current saturated when E/p was about 0.3 V/cm Torr. When the applied electric field was changed, no detectable decrease in light yield was observed at pressures below 3.0 atm for either component. Figure 3 shows a plot of the light yield of the slow component of the scintillation versus E/p .

The pulse shape of helium scintillation light was studied. Figure 4 shows oscillograms of the pulse at different pressures. The pulse shape consists of the slow component which extends to the order of 10^{-5} sec and the fast spike appearing on the leading edge of the pulse. The ratio of the number of emitted photons in the fast spike to that in the slow component was roughly estimated from the oscillograms to be 1:50. The typical characteristics of the fast spike observed at pressures greater than 2 atm were a rise time of 2×10^{-8} sec and a decay time of about 10^{-7} sec.

The shape of the slow component is well represented by the following formula:

$$C[\exp(-t/\tau_m) - \exp(-t/\tau_f)], \quad (1)$$

where C is a constant, t is the time in sec, and τ_f and τ_m are the rise and decay time constants, respectively, to be chosen to fit the shape. Figure 5 shows the variation of τ_f measured as a function of the pressure. The points represent the mean values obtained from several experimental runs. In the region of a pressure from 0.5 to 2.5 atm, τ_f was found to depend on the pressure in

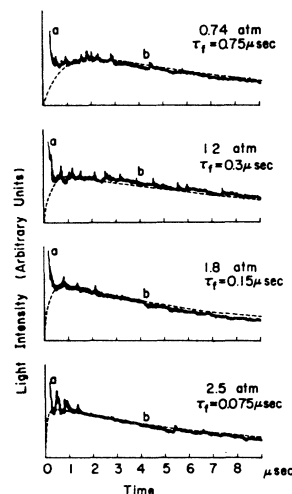


FIG. 4. Oscillograms of scintillation pulses induced by α particles at different helium gas pressures. Clipping time: 5×10^{-9} sec. a, fast spike; b, slow component. Decay time constant τ_m of the slow component is independent of the pressure within the accuracy of the measurements, and is estimated to be 10×10^{-6} sec. Dashed curve was calculated from Eq. (1) with two time constants τ_f and τ_m .

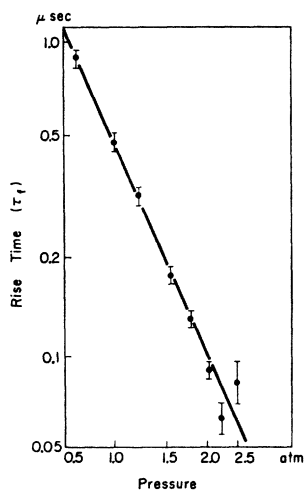


FIG. 5. Rise-time constant τ_f of the slow component of helium scintillation pulse as a function of the gas pressure. Least-squares analysis showed that τ_f was proportional to $p^{-2.2 \pm 0.3}$, where p is the helium gas pressure.

accordance with the following formula:

$$\tau_f = 0.3p^{-2.2 \pm 0.3}, \quad (2)$$

where p is expressed in Torr and τ_f is in sec. Note that τ_f was nearly proportional to the inverse square of the pressure. On the other hand, τ_m was independent of the pressure within the accuracy of the measurements and was about 10×10^{-6} sec.

When the sodium salicylate was covered with a lithium fluoride plate (40 mm diam and 0.7 mm thick), which is opaque to light of wavelength shorter than 1050 Å, the light yield of scintillation decreased markedly and the peak of the spectrum disappeared. The fast spike also disappeared from the oscilloscope traces. These facts indicate that almost no scintillation light is emitted in the wavelength region above 1050 Å.

B. Helium-Argon and Helium-Neon Mixtures

The scintillation yields of helium-argon and helium-neon mixtures were measured with a clipping time of 10×10^{-5} sec by using diphenyl stilbene. Also, the electric-field effect on the light yield was investigated. The pressure of the gas mixture was adjusted to maintain a constant α -particle range in the vessel. The range was 94% of the length between the source and the inner surface of the window. The pressure was 2 atm for helium and 0.39 atm for argon.

The light yields of helium-argon and helium-neon mixtures are shown in Fig. 6 as a fraction of the pressure of the heavier gas in the mixture. Normalization was made to the light yield of the pure helium. In helium-argon, the light yield decreases rapidly and then increases slowly with increasing concentration of argon. The minimum light yield was observed at an argon concentration of less than 1%. In helium-neon, the

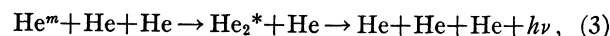
minimum light yield occurred at a neon concentration of about 20%, and the decrease was not so sharp as compared with that in the helium-argon mixture. These pronounced decreases are of the same nature as those observed by Northrop and Gursky, who used a clipping time of 0.5×10^{-6} sec. The decreases in the light yield of the helium-argon and helium-neon mixtures were also observed for the clipping time of 0.5×10^{-6} sec.

At any concentration of argon or neon in helium, the application of an electric field up to $E/p \sim 1.0$ V/cm Torr did not result in any detectable decrease in scintillation light yield for the clipping time of 10×10^{-5} and 0.5×10^{-6} sec (Fig. 3). The electric-field effect on the fast component was also studied, but no decrease in light yield was observed.

IV. DISCUSSION

The experiment on the electric-field effect showed that excited atoms, not recombination of ions, make an important contribution to the scintillation process of pure helium gas of less than 3 atm pressure when excited by α particles. We shall first discuss the slow component, which greatly contributes to the light yield of helium scintillation. Since the rise-time constant τ_f of the slow component is inversely proportional to the square of the gas pressure, a three-body collision should be involved in the scintillation process.

Thus we conclude that a metastable helium atom collides with two ground-state helium atoms to form an excited helium molecule (collision process). Subsequently this molecule deexcites, emitting a photon (radiation process). The process is shown in the following:



where He^m and He_2^* denote the metastable¹¹ atom and the excited molecule, respectively. The time constants τ_f and τ_m correspond, respectively, to the formation time and the lifetime of the excited molecule. The photon in process (3) is expected to have a wavelength

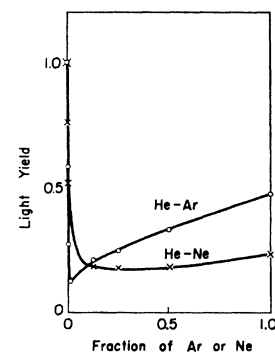


FIG. 6. Scintillation light yield of helium-argon and helium-neon mixtures for the clipping time of 10×10^{-5} sec normalized by that obtained for pure helium. Scintillation light yield obtained by using the clipping time of 0.5×10^{-6} sec showed nearly the same characteristics.

¹¹ Here, the metastable state means the 2^1S or 2^3S level of the helium atom. The 2^3P level is excluded because of its short lifetime ($\sim 10^{-7}$ sec). (See Ref. 4.)

in the far-ultraviolet region ($600\sim 1000\text{ \AA}$),¹² and is not absorbed by resonance. This was confirmed by an experiment that demonstrated that the scintillation light consisted of photons of wavelength less than 1050 \AA .

The collision process in (3) was suggested by Phelps and Molnar¹³ to interpret their experiment on helium afterglow by pulsed discharge. Colli¹⁴ and Forte¹⁵ have suggested that the same kind of collision process occurs in argon-gas scintillation. According to the report of Huffman *et al.*,¹⁶ the shape of the helium afterglow in the vacuum ultraviolet ($\sim 810\text{ \AA}$) region is similar to that of our observation. These findings support our conclusion that helium-gas scintillation can be mainly ascribed to process (3).

Process (3) is inconsistent with the proposal which Northrop and Gursky made for the interpretation of the scintillation of a binary gas mixture.¹ Their proposal was that the primary ions first form molecular ions by collision with normal atoms and then the molecular ions recombine with electrons emitting photons. Additional evidence that the recombination process was not important was given by the pulse-shape analysis. If the scintillation light of the helium gas is emitted through the recombination process, the decay time constant τ_m must depend on the gas pressure. The result shows that this is not the case.

Because the fast spike is not subject to the influence of an electric field, it cannot be attributed to ion recombination. This component may be ascribed to the light emitted from atomic resonance levels or the excited states of ions, as suggested by Northrop and Gursky.

In the helium-argon mixture, there should be competition between process (3) and the following quenching process (Penning process):



The cross section of process (4) is so large compared with that of the collision process in (3) that a mixing of very small amounts of argon into helium gas gives rise to the pronounced decrease in scintillation light. This interpretation is the same as that proposed by Jesse and Sadauskis¹⁰ for the pronounced decrease in W value (average energy for the production of an ion pair) for helium gas which is contaminated with very small amounts of argon or other gases, and is not consistent with the charge-transfer process as suggested by Northrop and Gursky.

Since the metastable helium atom can not ionize the neon atom energetically, the decrease in the light yield for the helium-neon mixture cannot be explained by the

¹² Y. Tanaka, Sci. Rep. I. P. C. R. **39**, 471 (1942); R. E. Huffman, Y. Tanaka, and J. C. Larrabee, J. Opt. Soc. Am. **52**, 853 (1962).

¹³ A. V. Phelps and J. P. Molnar, Phys. Rev. **89**, 1202 (1953).

¹⁴ L. Colli, Phys. Rev. **95**, 892 (1954).

¹⁵ M. Forte, Nuovo Cimento **3**, 1443 (1956).

¹⁶ R. E. Huffman, J. C. Larrabee, Y. Tanaka, and D. Chambers, J. Opt. Soc. Am. **55**, 101 (1965).

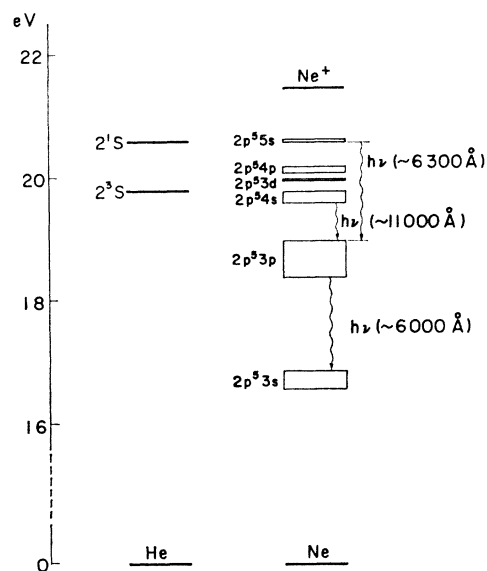
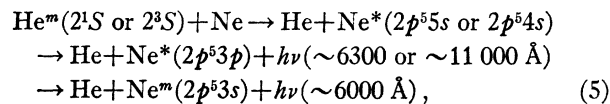


FIG. 7. The energy-level scheme of helium and neon for the explanation of the pronounced light-quenching effect in the helium-neon mixture in terms of the excitation transfer.

Penning process. Although the ionization of neon by an excited (not metastable) helium atom is energetically possible, Jesse and Sadauskis did not observe any significant decrease in W value with small amounts of neon added to helium. Thus the ionization process (4) is unlikely in the quenching process for the helium-neon mixture. From the viewpoint that neon has some excited levels in the neighbourhood of the metastable levels of a helium atom, we consider that the quenching process will be the following:



where Ne^* denotes the excited neon atom of comparatively short life and Ne^m the metastable neon atom. The experiment of Bennett,⁴ who observed the atomic spectra of inert gases at ordinary pressure excited by α particles, indicated that as a result of collisional energy transfer the atomic excitation accumulates in the states (for example, $2p^5 4s$) that are separated from the lower adjacent level by 0.5 eV or more. The wavelength of photons emitted from the excited neon atom in (5) is in the region where the spectral sensitivity of the photomultiplier is very low. Furthermore, the lifetime of the neon atom at the metastable level ($2p^5 3s$) is longer than the clipping time of our apparatus. This process is illustrated schematically in Fig. 7.

It is possible from the rise-time constant τ_f to make an estimate of the cross section σ_f for the formation of an excited helium molecule from a metastable helium atom through the collision process in (3). The cross section σ_f for the 0.5~2.5-atm region at 300°K is ob-

tained as follows:

$$\sigma_f = 50 \times 10^{-23} p (\text{Torr}) \text{ cm}^2.$$

We assumed that σ_f is obtained by $1/n_{\text{He}} V_f \tau_f$. Here τ_f is given by Eq. (2), and n_{He} is the number of helium atoms per cc and V_f is the relative velocity of approach for the interaction of the metastable atom with the ground-state atom.

Phelps and Molnar¹³ have measured the rate of destruction in the helium atom of the metastable state (2^3S) in the afterglow of pulsed discharge, and found that the cross section σ_f was $3 \times 10^{-23} p \text{ cm}^2$ for the 20~100 Torr region. This value agreed well with the value recently obtained by Benton *et al.*¹⁷ Both values are about 10 times smaller than our value. However, these values should not be directly compared with our results, because their cross sections are relevant to the destruction of $\text{He}(2^3S)$, while our cross section involves the reaction related to more highly excited states as well.

From studies of the density of metastable molecules by microwave pulsed discharge of helium gas, Phelps¹⁸ estimated that the natural lifetime of the $2^3\Sigma$ metastable molecular state was at least larger than 0.05 sec, and he concluded that the 2^3S metastable atoms were completely converted to $2^3\Sigma$ metastable molecules by the collision process in (3). In our experiments, the observed lifetime of excited helium molecules was about 10×10^{-6} sec, which is much shorter than that of the $2^3\Sigma$ state estimated by Phelps. This fact supports the inference that not only the 2^3S state but also the 2^1S state is involved in the formation of excited molecules by the metastable helium atom.

Jesse and Sadauskis¹⁰ interpreted the pronounced decrease in the W value of helium gas contaminated by small amounts of impurity as the result of competition between processes (3) and (4). The argon atom concentration C for which the probabilities of collision in both processes become equal is given by the following formula:

$$1/C = n_{\text{He}}/n_{\text{Ar}} = \sigma_i V_i / \sigma_f V_f, \quad (6)$$

where σ_i is the cross section for process (4), n_{Ar} is the number of argon atom per cc, and V_i is the relative velocity between metastable helium atoms and argon atoms. Using Eq. (6), Jesse and Sadauskis obtained experimentally the value of 9.3×10^3 for the ratio of σ_i to σ_f at a pressure of 875 Torr.

It is concluded that the competition between processes (3) and (4) is involved in the scintillation process of a helium-argon mixture. The ratio of the cross sections of the two processes can be estimated by the method of

Jesse and Sadauskis. Although the present experiment did not sufficiently cover the very low concentrations of argon, the observed light-yield curve agreed well with that of Northrop and Gursky. Thus, we estimated the ratio $\sigma_i/\sigma_f = 0.91 \times 10^3$ using the experimental result of Northrop and Gursky at a helium-gas pressure of 3000 Torr. Assuming that σ_f is proportional to the pressure and σ_i is independent of it, the ratio σ_i/σ_f at 875 Torr is obtained as 3.1×10^3 . This value roughly agrees with that of Jesse and Sadauskis. This fact supports our interpretation that the ionization process discovered by Jesse and Sadauskis for helium gas containing a small amount of argon can be applied to the gas scintillation process.

Using the ratio σ_i/σ_f and the cross section σ_f obtained by our experiments, we estimated the cross section of process (4). The cross section σ_i thus obtained is $41 \times 10^{-16} \text{ cm}^2$ for the ratio of Jesse and Sadauskis and $14 \times 10^{-16} \text{ cm}^2$ for the ratio of Northrop and Gursky. Benton *et al.*¹⁷ reported that the ionization cross section of argon due to the 2^1S metastable helium atom was $55 \times 10^{-16} \text{ cm}^2$, and that due to the 2^3S metastable atom was $6.6 \times 10^{-16} \text{ cm}^2$. They pointed out, however, that the cross section due to the singlet may be too high by a factor of as much as 2 or 3. The cross sections estimated by us are between the values as mentioned above. This fact is consistent with our interpretation that the 2^1S state rather than the 2^3S state gives the main contribution to the helium-gas scintillation process.

V. CONCLUSION

From experiments on scintillation in pure helium, helium-argon, and helium-neon mixtures excited by α particles, the following conclusions were drawn: (1) The recombination light is negligibly small at pressures less than 3 atm; (2) most of the photons from the pure helium have a wavelength of less than 1050 Å, and they are released from the decay of a metastable molecule formed by a three-body collision between a metastable and two ground-state atoms; (3) the metastable molecules seem to be formed from $\text{He}(2^1S)$ rather than from $\text{He}(2^3S)$; (4) the pronounced decrease in the light yield from helium with small amounts of argon is due to the Penning process; and (5) the pronounced decrease in the light yield from helium with small amounts of neon is due to the excitation transfer from helium to neon atoms.

ACKNOWLEDGMENTS

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¹⁷ E. E. Benton, E. E. Ferguson, F. A. Matsen, and W. W. Robertson, *Phys. Rev.* **128**, 206 (1962).

¹⁸ A. V. Phelps, *Phys. Rev.* **99**, 1307 (1955).