Evidence of Rare-Gas Quenching of Atomic Resonance Radiation

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The ratio of fluorescence intensities of the boron 2089- and 2498-Å lines is shown to depend on rare-gas pressure for both xenon and krypton. This is interpreted as evidence of rare-gas quenching of the boron resonance radiation.

The question of whether atomic resonance radiation can be quenched by the rare gases has been the subject of considerable speculation over the past fifty years.[1-10] Unlike molecular gases, the rare gases have no energy levels in a region suiable for transfer of the excitation energy, and hence for quenching to occur the full excitation energy (typically 2–6 eV) must be transferred into kinetic energy of the colliding atoms. Such a process is only possible if, during the collision, crossings or near-crossings occur between diabatic molecular-energy curves which correlate with the excited and ground states of the atom in question, or if a third curve crosses both excited and ground-state curves.[5,10]

In the early investigations of quenching, measurements were usually made by studying the intensity of the resonance radiation as a function of foreign-gas pressure; evidence of quenching was reported for the systems Hg/He,[1] Hg/Ar,[1] Na/He-Ne,[2] Na/Ar,[3] and Na/He.[4] It has since been pointed out, however, that in each of the above cases the observed quenching was probably not real and may have resulted from the effects of pressure broadening (and shift), radiation trapping, or molecular impurities.[7-10] More recently, several attempts have been made to detect rare-gas quenching by making precise measurements of atomic lifetimes,[9-11] but in each case no evidence of quenching was found; upper limits for the quenching cross sections were placed at 0.01 Å^2 for Na/Ne, Ne, Ar, Kr, and Xe,[9] at 0.001 Å^2 for Cs/He,[10] and at 0.002 Å^2 for Hg/Xe.[11] In addition, measurements of the reverse reaction to rare-gas quenching, i.e., atomic excitation produced by collisions with rare-gas atoms, have shown that the threshold energy for collisional excitation is much greater than the actual excitation energy for collisions between energetic potassium atoms and rare-gas atoms.[12] This indicates that the cross section for thermal quenching of potassium resonance radiation by the rare gases should be negligibly small.

In a recent paper describing some unusual properties of boron emission lines in glow-discharge lamps,[13] it was proposed that excitation of boron can result from low-energy collisions ($\approx$10 eV) between sputtered boron atoms and argon, krypton, or xenon atoms. Moreover, as the threshold energy for this excitation process appeared to be not much greater than the actual excitation energy, it was suggested that it might also be possible to observe the reverse (quenching) reaction at moderately low gas temperatures. In this Letter we report the results of a search for quenching of boron 2498-Å ($2s^2 3s^2 S_{1/2} - 2s^2 2p^2 P_{3/2,1/2}$) and 2089-Å ($2s 2p^2\,^2D_{5/2,3/2} - 2s^2 2p^2 P_{3/2,1/2}$) resonance radiation by neon, argon, krypton, and xenon at temperatures close to room temperature. For convenience we refer to each of these closely spaced multiplets as a single line.

Unfortunately, none of the currently available techniques for measuring atomic lifetimes appears to be suitable for studying the quenching of either the boron 2498-Å radiation ($\tau \approx 4$ ns) or the lower-wavelength 2089-Å radiation. We have therefore resorted to the traditional, but less reliable, method of measuring changes in the intensity of the resonance radiation with rare-gas pressure, making allowance for known sources of error. In addition, because of the difficulty of vaporizing boron by thermal methods, we have employed the technique of cathodic sputtering, which has previously been shown to be a suitable means of producing atomic vapors for studying resonance radiation.[14,15] In the case of boron, this method of vaporization is complicated by the presence of intense, abnormal (boron) emission from the sputtering discharge[13]; satisfactory signals could, however, be obtained by pulsing the discharge and using delayed detection, so that the resonance radiation was detected only after the abnormal boron emission had decayed away.

The sputtering-type resonance cell used in our investigation was a Pyrex glow-discharge lamp fitted with two pairs of silica windows.[15] The

cathode was a flat disk of boron, 1.2 cm in diameter. The rare gases used in the lamp were research-grade British Oxygen Company gases. The boron resonance radiation at 2498 and 2089 Å was detected sequentially by a wide-bandpass (60 Å) monochromator. The photomultiplier output was fed into a boxcar averager (PAR 162), in which the gate was positioned typically 10 ms after the sputtering pulse; this time interval allowed the emission in the discharge to decay to a tolerable level and was short enough that an adequate density of boron vapor remained in the observation region. The source used for excitation of the sputtered boron was a high-intensity hollow-cathode lamp,[16] which was filled with neon and operated in a high-current, pulsed mode in synchronism with the gate of the boxcar averager. For absorption measurements the position of the light source was changed, but all other conditions remained unaltered.

When the sputtering method of vaporization is used, alteration of the rare-gas pressure causes changes in the sputtering conditions, which can result in significant variations in the density of the atomic vapor and hence in the measured fluorescence signals. Attempts to correct the fluorescence signals using absorption measurements proved unsuccessful, because the changes in gas pressure also resulted in changes in the spatial distribution of the sputtered vapor, and these affected the fluorescence and absorption signals differently. Such problems do not arise, however, if the ratio of the 2089- and 2498-Å fluorescence signals, $R_F = F_{2089}/F_{2498}$, is studied, rather than the individual fluorescence signals. This ratio is independent of the atom density (in the region of low absorption), and is expected to be sensitive to the presence of quenching because of the differing lifetimes of the ^2D and $^2S_{1/2}$ excited states (24 and 4 ns, respectively[17]). However, allowance must be made for variations in the relative absorptive powers of the two lines at differing pressures. These variations, which amounted to typically 15% over the range of pressures investigated (5–55 Torr), include contributions due to differences in the effects of pressure broadening and pressure shifts on the two absorption lines.

Thus, for accurate measurements of quenching, it was necessary to study the quantity $R = R_F/R_A$, where $R_A = A_{2089}/A_{2498}$ is the ratio of the absorptions. For the measurements of R_A, the density of the boron vapor was adjusted so that the 2498- and 2089-Å absorptions were typically 10% and 4%, respectively. For the measurements of R_F, the absorptions were kept to less than 2% to eliminate effects of radiation trapping. (At this low level of absorption, R_F was found to be independent of the density of boron vapor.) A small correction (< 5%) was required to convert the measured value of R_A to a value corresponding to the lower-absorption conditions used in the fluorescence measurements.

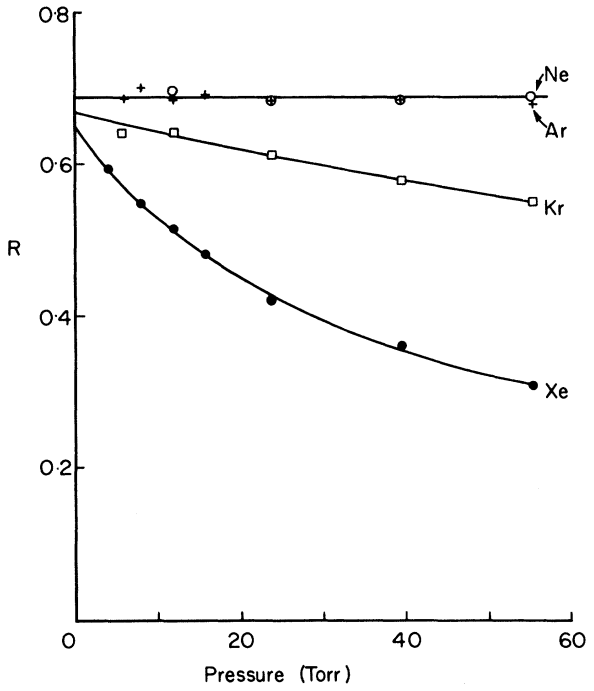

FIG. 1. Pressure dependence of the corrected ratio ($R = R_F/R_A$) of the intensity of boron 2089-Å to boron 2498-Å resonance radiation for various rare gases. The solid lines represent a fit of Eq. (1) to the experimental points.

Figure 1 shows the quantity $R = R_F/R_A$ plotted as a function of gas pressure for neon, argon, krypton, and xenon. The curves for neon and argon are found to remain constant with pressure, whereas with xenon, and to a lesser extent krypton, there is a definite pressure dependence, which we attribute to quenching, mainly of the 2089-Å resonance radiation. The small differences in the intercepts at zero pressure for the four curves are attributed to cumulative errors in the quantity R, which are larger between different sets of measurements (different gases) than within a given set of measurements (same gas).

The ratio R can be expressed as the quotient of two Stern-Volmer[7] expressions, viz.

$$R = R_0(1 + pC_{2498})/(1 + pC_{2089}), \quad (1)$$

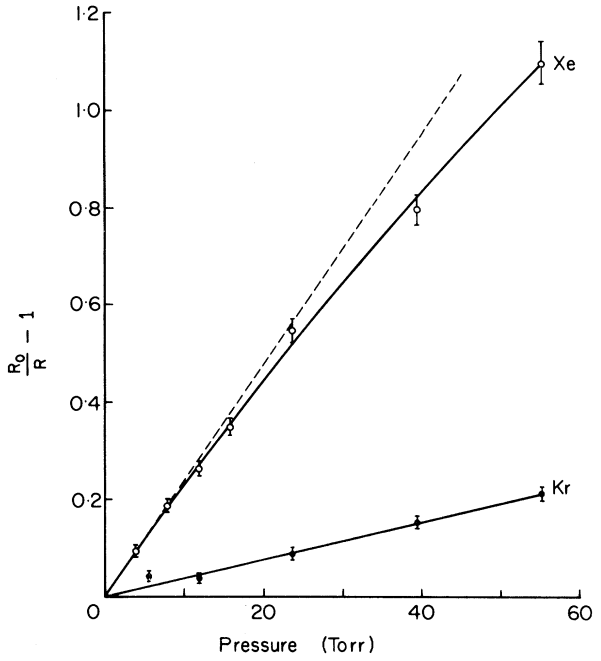

FIG. 2. Pressure dependnece of $(R_0/R) - 1$ for Xe and Kr.

where R_0 is the value of R at zero pressure, $C = Q\tau\bar{v}/kT$, Q is the effective quenching cross section at temperature T, τ is the radiative lifetime of the excited state, and $\bar{v}$ is the mean relative velocity of the colliding atoms. In the absence of of quenching of the 2498-Å radiation ($C_{2498} = 0$), Eq. (1) predicts that a plot of $(R_0/R) - 1$ versus gas pressure should yield a straight line passing through the origin. The curve obtained in the case of xenon (Fig. 2) shows some curvature at the higher pressures, suggesting the presence of a small amount of quenching of the 2498-Å radiation. An analysis of the data, using Eq. (1), yields the values $C_{2089}(\mathrm{Xe}) = 0.027\ \mathrm{Torr}^{-1}$, $C_{2498}(\mathrm{Xe}) = 0.004\ \mathrm{Torr}^{-1}$, and for the (linear) krypton data, $C_{2089}(\mathrm{Kr}) - C_{2498}(\mathrm{Kr}) = 0.004\ \mathrm{Torr}^{-1}$. Assuming lifetime values of 24 and 4 ns for the $^2S_{1/2}$ and ^2D states,[17] respectively, we obtain for the effective quenching cross sections (at about 400 K) $Q_{2089}(\mathrm{Xe}) = 5 \pm 1$ Å^2, $Q_{2498}(\mathrm{Xe}) = 4 \pm 2$ Å^2, and $Q_{2089}(\mathrm{Kr}) = 0.9 \pm 0.2$ Å^2, assuming $Q_{2498}(\mathrm{Kr}) \leq Q_{2089}(\mathrm{Kr})$.

To check our interpretation of the observed pressure dependence of R, several tests were carried out. First, the measurements were repeated using several different bottles of research-grade xenon from different batches. These all gave the same results, suggesting that the observed quenching does not result from molecular impurities in the xenon. Second, measurements were made using variable amounts of xenon in a xenon-krypton mixture, in which the total gas pressure was kept constant. The results obtained, after allowing for the small quenching effect of the krypton, were the same as in Fig. 1, indicating that the observed reduction in R does not result from changes in the spatial distribution of the boron vapor with gas pressure. Finally, in the interpretation of the data, it has been a assumed that the observed pressure dependence of R results mainly from a reduction in F_{2089} (together with a small reduction in F_{2498}), rather than from an increase in F_{2498}, or a combination of both effects. The only apparent process which could lead to an increase in F_{2498} is collisional transfer of excitation from the ^2D state to the $^2S_{1/2}$ state. This possibility was tested by using pure 2089-Å radiation to excite the boron vapor and looking for fluorescence at 2498 Å. A filter consisting of a solution of anthracene in pure ethanol was used to separate the 2089-Å radiation from the intense 2498-Å radiation emitted from the source. For all four gases at pressures up to 55 Torr, the ratio F_{2498}/F_{2089} was found to be less than 5%, which is much smaller than the observed reduction in R (50% for 55 Torr of Xe).

To our knowledge these results for B/Xe and B/Kr represent the first definite evidence of thermal quenching of resonance radiation by rare gases. They also provide further evidence for the occurrence of curve crossings at large internuclear separation in some boron–rare-gas systems.[13] The occurrence of these crossings in the thermally accessible (flat) region of the excited-state curves for xenon and krypton, but not argon, may be a consequence of the relative magnitudes of the dipole polarizabilities of the rare gases. In the case of argon, the crossing point apparently can be reached in collisions with energetic sputtered boron atoms (as in excitation[13]), but not in thermal collisions (as in quenching).

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Rapid Thermal Transport from Turbulent Skin Layer to Plasma Core in a Toroidal Experiment

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Anomalously rapid thermal transport from a skin layer to the plasma core has been observed in a toroidal turbulent heating experiment. The thermal transport rate is approximately two orders of magnitude faster than the classical rate based on the anomalous electron collision frequency.

Skin effects in toroidal turbulent heating experiments have been reported by several groups.[1-3] However, little is known of the transport of energy created in the skin layer, whether it is lost to the vacuum wall or it penetrates into the plasma core. In proposed schemes for turbulently heating a large toroidal plasma,[4] such as a reactor-size tokamak, one crucial assumption is that the thermal energy deposited in the skin layer is rapidly transported toward the plasma core. Otherwise, the electron temperature in the skin layer would become undesirably high, prematurely quenching current-driven instabilities. Although the assumption is based on some experimental facts previously reported, these were obtained in plasmas with a skin layer artificially created by ring electrodes[5] or a nonuniform electric field.[6]

In this Letter, we report experimental observations of rapid thermal transport from a genuine current skin layer toward the plasma core during a time interval when energy loss is still unimportant. The experiment is performed in a toroidal device described previously.[6,7] The mode III operation, or tokamak mode, is employed. A helium plasma with an electron density $(5-10)\times10^{13}$ cm^{-3} is prepared by rf preionizer and preheater pulses in a toroidal magnetic field of 3 kG. A toroidal electric field with a quarter period of 2.0 μs is inductively applied by discharging a 2.5 μF, 50 kV capacitor. Diagnostic methods include a 76-GHz microwave interferometer[8] for the electron density measurements, a small movable magnetic probe for measuring the local magnetic field (both poloidal and toroidal components), a small Rogowsky coil for direct measurement of the local current density, and a diamagnetic Rogowsky coil wound around the feeder cable to the toroidal magnetic field coils. The local magnetic field measurements yield a plasma current-density profile and a plasma pressure profile, and the diamagnetic Rogowsky coil measures the average plasma pressure. The total energy input into the plasma can be calculated from the total plasma current (measured by a Rogowsky coil) and the output voltage from a one-turn loop corrected for the plasma inductance.[9]

Figure 1 shows typical examples of the radial distribution of the poloidal magnetic field B_p and the change (increase) in the toroidal magnetic field ΔB_φ, observed at different times in the discharge, for $n_e\simeq1\times10^{14}$ cm^{-3}. [As shown later, in Fig. 4, β_p (poloidal beta) never exceeds unity and the plasma stays paramagnetic—hence the observed increase in the toroidal magnetic field.] The toroidal current density j_z can be calculated from $j_z\simeq\partial(rB_p)/r\partial r$ and is in agreement with direct measurements made with use of the small Rogowsky coil (Fig. 2). It is clearly seen from Figs. 1 or 2 that the plasma current is limited to a skin layer until about 1.5 μs after applying the main heating field. During this time interval, the distribution of the current layer is stable. After 1.5 μs the skin current penetrates into the column center within 0.3–0.5 μs.

The profile of the plasma pressure $P_\perp=n(T_e+T_i)$ over the cross section of the plasma column