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of

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G. Copley and L. Krause

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The Trapping and Quenching of Potassium Resonance Radiation

G. Copley² and L. Krause Department of Physics, University of Windsor, Windsor, Ontario

ABSTRACT

The effects of imprisonment of resonance radiation in potassium vapor and of collisions between excited potassium atoms and N₂, H₂, HD and D₂ molecules, on the measured lifetimes of the $4^{2}P$ states, have been investigated by the method of delayed coincidences. The potassium atoms were excited with short pulses of resonance radiation, and the delays between the exciting and fluorescent pulses were measured with a time-to-amplitude converter. At pure potassium densities below 10^{10} atoms/cm³, the measured lifetimes approach the constant values 27.8 ± 0.3 and 27.6 ± 0.8 nsec, corresponding to the natural lifetimes of the $4^{2}P_{1/2}$ and $4^{2}P_{3/2}$ states, respectively. At higher densities the effective lifetimes are in satisfactory agreement with the predictions of Holstein's theory. The decrease in the measured decay rates observed upon addition of the diatomic gases, yielded cross sections for quenching collisions with N₂, H₂, HD and D₂, which equal 34.0 ± 2.0 8^{2} , 9.4 ± 1.3 8^{2} , 11.9 ± 1.5 8^{2} and 8.0 ± 2.0 8^{2} , respectively.

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1. INTRODUCTION

When potassium vapor at low density is illuminated with potassium resonance radiation, some of the atoms are excited to the $4^{2}P_{1/2}$ or $4^{2}P_{3/2}$ state and subsequently decay emitting resonance fluorescence. The decay process, which is assumed to be exponential, is described by the equation

(1)
$$N_t = N_o \exp(-t/\tau)$$
,

where N_t and N_o are the densities of excited atoms at times t and 0, respectively, and τ , the natural lifetime of the excited state, is a specific property of the potassium atoms. If the vapor density is increased, the fluorescent radiation becomes imprisoned in the vapor, and measurements of the decay rates yield effective lifetimes, τ^{1} , which are significantly larger than τ and which are a property not only of the isolated atoms but also of the bulk vapor.

The trapping of resonance radiation in metallic vapors, and its effects, are of fundamental interest to workers in the fields of atomic physics, radiofrequency spectroscopy, upper atmospheric physics and plasma physics.

The imprisonment of resonance radiation in sodium was first observed by R. W. Wood (1905) and one of the early quantitative studies of the effect was reported by Zemansky (1922) who measured the lifetime of the Hg $6^{3}P_{1}$ state in relation to the mercury vapor pressure. The first systematic investigation of radiation trapping in sodium was carried out by Kibble, Copley and Krause (1967), although other authors (for instance, Seiwert, 1956) had used various existing theories of the effect to correct fluorescent

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intensity measurements.

Milne (1926) proposed the first comprehensive and quite successful theoretical treatment which employed a diffusion analogy as a model for the imprisonment of radiation. Holstein (1947, 1951) carried out a more complete analysis for a single resonance line, in which he also considered geometrical effects, and which was extended by Walsh (1959) to account for the influence of simultaneous Doppler and collision broadening and of hyperfine structure. When the hyperfine structure extends beyond the Doppler width of the resonance line, coherence may be preserved in the scattering of a photon from atom to atom (Barrat, 1959; D'Yakonov and Perel, 1965).

The addition of a molecular gas complicates the decay spectrum as inelastic collisions between an excited potassium atom and a molecule cause the atom to be transferred either to the ground state (quenching collisions) or to the other ^{2}P state (mixing collisions), with an accompanying change in the molecular translational, vibrational and rotational energy. Quenching competes with spontaneous decay and decreases the observed lifetime. ^{2}P mixing, which has been investigated in mixtures of alkali vapors and diatomic gases by McGillis and Krause (1968), can also affect the decay spe trum.

The early experiments involving quenching of atomic resonance radiation have been summarized by Pringsheim (1949). The quenching of potassium resonance radiation by diatomic molecules has been investigated by Jenkins (1968), Hooymayers and Alkemade (1966), Gatzke (1963), Smith, Stewart and Taylor (1954) and McGillis and Krause (1968). The knowledge of quenching cross sections obtained under well-defined experimental conditions should help in the understanding of the mechanism involved in the interactions

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between excited atoms and various molecules.

2. THEORETICAL

(a) Imprisonment of Radiation

The theories of Barrat (1959) and of D'Yakonov and Perel (1964) cannot be applied in the case of potassium, because the hyperfine structures of the resonance lines lie totally within their Doppler widths. Holstein's (1947, 1951) theory of radiation trapping should, however, provide a useful basis for comparison between experimental results and theoretical predictions.

Holstein's model involves a dimensionless 'escape factor', g, which appears in the exponential decay equation:

(2)
$$N(t) = N_0 \exp(-t/\tau^2) = N_0 \exp(-gt/\tau) ,$$

where g, a constant characteristic of the decay process, is evaluated from integro-differential equations which give the population of a state $n(\vec{r},t)$ in relation to position and time, and which are formulated in terms of the transmission coefficient for a Doppler-broadened line. The transmission coefficient, $T(\rho)$, is defined by the following general expression:

(3)
$$T(\rho) = \int P(\gamma) \exp[-k(\gamma)\rho] d\gamma$$

where $P(\gamma)$ represents the frequency spectrum of the radiation emitted from a given volume element and $k(\gamma)$ is the absorption coefficient which varies with the frequency γ . Thus, $T(\rho)$ is the probability that an emitted photon will traverse a distance $\rho = |\vec{r'} - \vec{r}|$ without being absorbed. If the shape

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of the (non-self-reversed) resonance line is determined by Doppler- or pressure-broadening, and the emitting atoms have a Maxwellian velocity distribution, then $P(\gamma) \propto k(\gamma)$. For a Doppler-broadened line (pressurebroadening may be neglected at very low vapor pressures), the transmission coefficient is expressed as:

(4)
$$T(\rho) = \int_{-\infty}^{\infty} (1/\pi^{1/2}) \exp(-x^2) \exp[-k_0 \rho \exp(-x^2)] dx$$
.

 k_0 is the absorption coefficient at the centre of the line of wavelength λ_0 (or frequency γ_0), and is given by

(5)
$$k_0 = (\lambda_0^3/8\pi) N(g_1/g_0) (\pi^{1/2} v_0 \tau)^{-1}$$

where $\mathbf{v}_0 = (2RT/M)^{1/2}$, is the average speed of the potassium atoms, N is their density and \mathbf{g}_1 and \mathbf{g}_0 are the respective statistical weights of the excited and ground states. The variable x in Eq. (4) is defined as $\mathbf{x} = (\gamma - \gamma_0)_{\mathrm{TC}}/\mathbf{v}_0$. The escape factor, g, is related as follows to $\mathrm{T}(\rho)$:

$$(6) g \simeq T(f)$$

where 2 is the shortest linear dimension of the fluorescence cell. Eq. (6) constitutes a good approximation and, together with Eq. (2) provides the connection between the theoretical and the observed decay spectra.

(b) The Decay of the Resonance States in the Presence of Quenching Collisions The processes which occur when a mixture of potassium vapor and

molecular gas is irradiated with potassium resonance radiation, are depicted in Fig. 1. The collision numbers Z_{10} , Z_{20} , Z_{12} and Z_{21} , are defined as the frequencies of inelastic collisions per excited atom, leading to the various

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processes of energy transfer. s_1 and s_2 are the numbers of potassium atoms excited per second to the appropriate 2p state. Since the excitation proceeds by a short pulse of light, s_1 and s_2 are time-dependent and, for the purpose of analysis, will be considered as step functions having the values s_1 and s_2 in the interval $0 \le t \le t_0$ and the value zero elsewhere. $1/\tau_1'$ and $1/\tau_2'$ are the observed decay rates of the 2p states in the absence of the gas. The following are the rate equations for the populations of the two resonance states.

(7)
$$\frac{dN_1}{dt} = s_1 + N_2 z_{21} - N_1 [1/\tau_1' + z_{10} + z_{12}] = s_1 + N_2 z_{21} - N_1 r_1',$$

(8)
$$\frac{dN_2}{dt} = s_2 + N_1 z_{12} - N_2 [1/\tau_2 + z_{20} + z_{21}] = s_2 + N_1 z_{12} - N_2 r_2'$$

Equations (7) and (8) have solutions of the form:

(9)
$$N_1 = A_1 \exp(-r_1 t) + A_2 \exp(-r_2 t) + A_3$$
,

(10)
$$N_2 = B_1 \exp(-r_1 t) + B_2 \exp(-r_2 t) + B_3$$

where

(11)
$$A_3 = (Z_{21}s_2 + s_1 f_2)/(f_1 f_2 - Z_{12}Z_{21}) ,$$

(12)
$$B_3 = (2_{12}s_1 + s_2 r_1)/(r_1 r_2 - 2_{12} r_{21}),$$

(13)
$$r_1 = (1/2)(\Gamma_1 + \Gamma_2) + (1/2)[(\Gamma_1 - \Gamma_2)^2 + 4z_{12}z_{21}]^{1/2}$$
,

(14)
$$r_2 = (1/2)(\Gamma_1 + \Gamma_2) - (1/2)[(\Gamma_1 - \Gamma_2)^2 + 4z_{12}z_{21}]^{1/2}$$
.

Equations (13) and (14) correspond to the solutions found by Bennet, Kindlmann and Mercer (1965). After excitation ceases at $t = t_0$, $s_1 = s_2 = 0$,

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yielding the initial conditions for decay:

(15)
$$\frac{dN_{10}}{dt} = N_{20}Z_{21} - N_{10}\Gamma_{1}$$
; $\frac{dN_{20}}{dt} = N_{10}Z_{12} - N_{20}\Gamma_{2}$

where $N_{10} = N_1(t_0)$ and $N_{20} = N_2(t_0)$ are obtained from Eqs. (9) and (10). Putting $t_0 = 0$, to eliminate an arbitrary factor, Eqs. (15) give:

(16)
$$A_1 = [N_{20}Z_{21} + N_{10}(r_2 - \Gamma_1)]/(r_2 - r_1) ,$$

(17)
$$A_2 = [N_{20}Z_{21} + N_{10}(r_1 - r_1)]/(r_1 - r_2) ,$$

(18)
$$B_1 = [N_{10}Z_{12} + N_{20}(r_2 - r_2)]/(r_2 - r_1) ,$$

(19)
$$B_2 = [N_{10}Z_{12} + N_{20}(r_1 - r_2)]/(r_1 - r_2) .$$

A useful simplification results if it is postulated that $Z_{10} \simeq Z_{20} = Z_{10}$ and that the square-root term can be reduced as follows.

$$\left[\left(\Gamma_{1}^{\prime} - \Gamma_{2}^{\prime}\right)^{2} + 4z_{12}z_{21}\right]^{1/2} = (z_{12}^{\prime} + z_{21}^{\prime}) \left\{1 + \left[\left(1/\tau_{1}^{\prime} - 1/\tau_{2}^{\prime}\right)/(z_{12}^{\prime} + z_{21}^{\prime})^{2}\right]\right]$$
$$\left[1/\tau_{1}^{\prime} - 1/\tau_{2}^{\prime} + 2(z_{12}^{\prime} - z_{21}^{\prime})\right]^{1/2} \simeq z_{12}^{\prime} + z_{21}^{\prime} \cdot$$

The first assumption has been verified experimentally in mixtures of alkali vapors and diatomic gases by McGillis and Krause (1968) and by Belisio, Davidovits and Kindlmann (1968). The second approximation is accurate to better than 3_{p} at the potassium vapor pressure (10^{-6} Torr) and gas pressures (1 - 3 Torr) used in this investigation. With these approximations, r_1 and r_2 become

(20)
$$r_{1} = (1/2)(1/\tau_{1}' + 1/\tau_{2}') + Z_{12} + Z_{21} + Z_{12} + Z_{1$$

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At low potassium vapor pressures, when $1/\tau_1^{\dagger} \simeq 1/\tau_2^{\dagger}$, $A_1 = -B_1$. When both resonance components are observed at the same time so that

(21)
$$N(t) = N_1(t) + N_2(t),$$

the decay spectrum is determined to a good approximation by r_2 alone, and the decay equation becomes

(22)
$$N(t) = (A_2 + B_2)exp(-r_2t)$$

Equation (22) treats the $4^{2}P_{1/2}$ and $4^{2}P_{3/2}$ states as a single resonance state, with the spontaneous decay rate $(1/2)(1/\tau_{1}^{1} + 1/\tau_{2}^{1})$ and radiationless decay (quenching) rate Z. The latter yields the quenching cross section, Q, through the equation

where N is the density of the quenching molecules and v_r is the average relative velocity of the colliding partners.

3. EXPERIMENTAL

The apparatus and experimental procedure were similar to those used previously for experiments with sodium atoms (Kibble, Copley and Krause 1967ab; Habib, Kibble and Copley 1968). Because of the longer potassium resonance wavelengths and longer lifetimes of the 4²P states, minor modifications were made to the pulsing and detection systems and to the electronic circuits (Copley 1968).

The excitation of the potassium atoms was accomplished by potassium

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resonance radiation from an Osram spectral lamp, which was resolved into its two components with an interference filter, was brought to a focus between the plates of a Kerr shutter and again inside a fluorescence cell containing potassium vapor. The shutter was opened by a 20 or 50 nsec highvoltage pulse, a sample of which, shaped to a rectangular pulse of 200 nsec duration, was applied to one input of a time-to-amplitude converter. Fluorescent photons emitted by the potassium atoms were detected with a photomultiplier whose output pulses, after shaping, were applied to the other input of the time-to-amplitude converter, producing a ramp pulse of amplitude proportional to the time overlap between the two input pulses. The ramp pulse, also suitably shaped, was accumulated in a kicksorter channel appropriate to its amplitude, where it became part of the resulting spectrum of decay times. The kicksorter (Victoreen PIP 400) was gated to reject pulses of too small amplitude and improper shape.

The fluorescence cell and its side-arm were mounted in an oven which maintained the temperature of the main cell constant within $\pm 1^{\circ}$ C, and the somewhat lower temperature of the side-arm, which contained liquid potassium, within $\pm 0.2^{\circ}$ C. During the experiments with the Vapor-gas mixtures, the main body of the cell was maintained at $80 \pm 1^{\circ}$ C. The gases^{*} were purified by gettering with hot alkali vapor (Xibble, Copley and Krause 1967b) and their pressures were measured with a trapped McLeod gauge.

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^{*} Linde M.S.C. grade H₂ and H₂ of nominal purity 99.99%, HD (99%) supplied by Stohler Isotope ChemicalS, and Matheson C.P. grade D₂ (99.5%) were used in the experiments.

4. RESULTS AND DISCUSSION

(a) Imprisonment of Radiation

1g. 2.

A typical decay spectrum, containing a total of about 5×10^4 counts accumulated in two hours, is shown in Fig. 2. The slight undulations about the straight line which represents a strictly exponential behaviour of the decay, are due partly to statistical error and partly to the imperfect shape of the sample pulse taken from the Kerr cell. The spectrum also contains some background arising from light leaking through the shutter and from the small dark current in the photomultiplier, which amounted to 13 counts/channel, uniformly distributed in time. This background was subtracted before subjecting the results to further analysis which was carried out on an I.B.M. 1620 (II) computer and which yielded the apparent lifetimes τ_1^i and τ_2^i .

1g. 3.

Fig. 3 shows the apparent lifetimes which were determined over a range of potassium vapor pressures. At low densities, τ_1^{\dagger} and τ_2^{\dagger} approach the limiting values τ_1 and τ_2 which are very nearly equal to one another. As the vapor pressure increases, τ_1^{\dagger} and τ_2^{\dagger} rise steeply with the curves separated by a factor of two on the pressure scale which, as expected, equals the ratio of statistical weights of the ^{2}P states. The individual values τ_1^{\dagger} and τ_2^{\dagger} are subject to experimental error of 1% arising from the uncertainties in the background count and from the possibility of 'pile up' which was reduced to a minimum by keeping the intensity of the exciting light so low that, on the average, fewer than one fluorescent photon was observed for each ten exciting light pulses. A correction made for this effect was negligible at low values of τ' but amounted to as much as 20%

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for values $\tau^{\dagger} > 10\tau$. The effect on τ^{\dagger} of small changes in the depth at which fluorescence was observed was found to be negligible.

The solid lines in Fig. 3 represent Eq. (6), fitted at the encircled point by assuming l = 5.5 mm and $\tau_1 = \tau_2 = \tau = 27.7$ nsec. The agreement with the experimental results is good except in the elbow of the curve where $k_0 l \simeq 1$. The value l = 5.5 mm corresponds directly to the Kerr cell spacing of 6 mm which, as a stop in the optical system, determines the depth of fluorescing vapor through 1:1 imaging. The slight lack of agreement in the elbows of the curves is probably due to the fact that Holstein's theory assumes a Maxwellian velocity distribution in the potassium vapor, which is not necessarily correct (Holstein 1947).

To obtain the natural lifetimes τ_1 and τ_2 , it was necessary to determine the pressure at which radiation trapping becomes noticeable. It was found that plots of fluorescent intensity against pressure became non-linear at 3.5 x 10⁻⁷ and 7.0 x 10⁻⁷ Torr, for the $4^2P_{3/2}$ and $4^2P_{1/2}$ states, respectively. The values τ_1 and τ_2 , listed in Table I, were obtained as weighted averages of the lifetimes measured at pressures lower than these. Even though lack of sufficient intensity made it impossible to make measurements at pressures below 10^{-7} Torr, the results are in good agreement with recent values of other authors. The discrepancy with Ostrovskii and Penkins' result (1962) is ascribed to the fact that the look method depends on the exact knowledge of the potassium vapor pressure which is difficult to measure with high precision.

(b) The Quenching Cross Sections

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The decay spectra obtained at various pressures of the quenching

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gases were exponential in character and appeared similar to those observed with pure potassium vapor: No correction for pileup to the measured value r_2 (the characteristic decay rate in Eq. (21)) was necessary, because the time constant $1/r_2$ was short and the counting rate low. τ_1^{\dagger} and τ_2^{\dagger} , which appear in the equation

(24)
$$Z = r_2 - (1/2)(1/\tau_1^{\dagger} + 1/\tau_2^{\dagger})$$

were determined before each experimental run and were found to remain constant within $\pm 3\%$ at a given potassium vapor pressure.

The quenching of the fluorescence by N_2 , H_2 , HD and D_2 is represented in Fig. 4. As expected, the plots of ZT, the number of effective quenching S. 4. eollisions per lifetime of an excited atom, against gas density N, are linear and pass through the origin. Most measurements were made at the potassium vapor pressure of 1×10^{-6} Torr and with the 7665 Å resonance component used for excitation, which afforded the best comprehise between adequate intensity of signal and the requirement that $\tau_1^! \simeq \tau_2^!$. Some measurements were also carried out at a vapor pressure of 4 x 10^{-7} Torr, or using the 7699 $\frac{9}{4}$ component for excitation, to ensure that neither a possible change in τ_1^+ or τ_2^+ due to broadening collisions nor a decrease in density of ground state atoms upon addition of the quenching gases, was a source of systematic error. The results of all the experimental runs yielded straight lines whose slopes were equal, within experimental error, to those shown in Fig. 4. The quenching cross sections obtained from the slopes, are compared in Table II with values reported by other authors. le II The quoted errors which arise from counting statistics, were calculated from a least squares analysis of the experimental results. The values

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reported by Nooynayers and Alkemade (1965) and by Jenkins (1968), obtained from investigations of flames, as well as Gatzke's (1953) cross sections from the photolysis of KI, were determined at quite high relative velocities v. It is not really surprising that these cross sections are lower than those obtained from intensity measurements by Smith, Stewart and Taylor (1954), by McGillis and Krause (1968) as a by-product of excitation transfer studles, and in this investigation. It was not possible to separate the individual cross sections $Q_{1,0}$ and $Q_{2,0}$, because the numerical procedures by which the decay constants are extracted from the available experimental data (each measurement yields the sum of two exponentials), are not sufficiently accurate, nor can their inherent accuracy be improved (Rogers 1962). The agreement with the results of McGillis and Krause (1968) is quite reasonable, except for D₂. There is no apparent correlation of the quenching cross sections with the energy intervals between the potassium resonance levels and the mearest vibrational levels in the molecules. A similar lack of correlation has been noted by Starr (1955) who investigated the transfer of energy from excited N₂ molecules to ground-state sodium atoms. It is possible that such a resonance effect, if present, might have been masked by the variation of the cross sections with the relative velocity which was different for all the molecules. More experimental avidence will have to be obtained before any valid conclusions concerning the presence of such resonances can be reached.

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TABLE I

Lifetimes of the 4²P States in Potassium

τ (nsec)				
4 ² P _{1/2}	4 ² P3/2	Source	Method	
27.8 <u>+</u> 0.8	27.6 <u>+</u> 0.8	This Investigation	Delayed coincidences	
-	26.0 <u>+</u> 0.5	Schmieder, Lurio and Happer (1968)	Level crossing	
27.8 <u>+</u> 0.5	27.8 <u>+</u> 0.5	Link (1966)	Phase shift	
25.9 <u>+</u> 0.8	25.9 <u>+</u> 0.8	Ostrovskii and Penkin (1962)	Hook method	
27.1	26.9	Heavens (1961)	Bates-Damgaard calculation	
27.1 <u>+</u> 0.9	27.1 <u>+</u> 0.9	Stephenson (1951)	Magneto-rotation	

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TABLE II

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Cross Sections for Quenching of Potassium Resonance Radiation

C	Cross Sections for Collisions with Molecules $\binom{0^2}{n}$			
Source	K - N ₂	K - H ₂	K - HD	K - D ₂
This Investigation	34 . 0 <u>+</u> 2.0	9.4 <u>+</u> 1.3	11.9 ± 1.5	8.0 <u>+</u> 2.0
McCillis and Krause (1963) - Q ₁₀	35.0 🛨 7.0	7.0 ± 3.0	11.0 ± 4.0	2.0 ± 1.0
McGillis and Krause (1968) - Q ₂₀	39.0 <u>+</u> 8.0	4.0 <u>+</u> 1.5	14.0 <u>+</u> 3.0	1.0 ± 0.5
Jenkins (1963)	17.6 ± 0.9	3.2 <u>3+</u> 0.16	-	-
Nooymayers and Alkemade (1966)	29 . 5 <u>+</u> 1.0	5.4 ± 0.3	-	-
Gazzke (1963)	20 . 2 <u>+</u> 2.0	-	-	-
Smith, Stewart and Taylor (1954)	-	ó.2	-	4.5

FIGURE CAPTIONS

- Fig. 1. Energy levels involved in sensitized fluorescence and in the quenching of potassium resonance radiation. The solid arrows represent optical excitation and spontaneous decay, the broken lines refer to collisional processes.
- Fig. 2. A semilogarithmic plot of the decay spectrum in pure potassium vapor at a pressure of 1.3 x 10^{-6} Torr, accumulated during two hours, using 7665 Å excitation. The slope yields $\tau_2^1 = 43.3$ nsec.
- Fig. 3. The variation of τ_1^{\dagger} and τ_2^{\dagger} , with potassium vapor pressure. $\tau = 27.7$ nsec and the encircled point on the ${}^{2p}_{3/2}$ curve, were used to fit the theoretical curves, representing Holstein's relation $1/g = \tau'/\tau$, to the experimental results.
- Fig. 4. Quenching of potassium fluorescence by N_2 , H_2 , HD and D_2 . The error bars represent typical statistical errors which are nearly the same for all the points. ∇ , N_2 ; o, H_2 ; +, HD; x, D_2 . $\tau = 27.7$ nsec.

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Copley and Krause Fig. 1



Copley and Krause Fig. 2



Copley and Krause Fig. 3





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and of collisions between excited j	potassium atoms an	a_{N_2} , a_{2} , a_{2} , a_{2} and b_{2}			
molecules, on the measured lifetimes of the $4^{\mathbf{z}}\mathrm{P}$ states, have been investigated					
by the method of delayed coincidences. The potassium atoms were excited					
with short pulses of resonance radiation, and the delays between the exciting					
and fluorescent pulses were measured with a time-to-amplitude converter.					
At pure potassium densities below 10^{10} atoms/cm ³ , the measured lifetimes					
approach the constant values 27.8 ± 0.8 and 27.6 ± 0.8 nsec. corresponding					
to the natural lifetimes of the $4^{2}P$, and $4^{2}P$ states respectively. At					
higher densities the affective lifetimes are in Statisfactory accompt with					
the ending of the effective three are in satisfactory agreement with					
the predictions of Holstein's theory. The decrease in the measured decay					
rates observed upon addition of the diatomic gases, yielded cross sections					
for quenching collisions with N ₂ , H ₂ , HD and D ₂ , which equal 34.0 \pm 2.0 Å ² ,					
9.4 \pm 1.3 Å ⁺ , 11.9 \pm 1.5 Å ⁺ and 8.0	0+ 2.0 X, fespec	tively.			

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