

LIFETIMES OF EXCITED LEVELS OF NEUTRAL
AND IONIZED STRONTIUM BY THE HANLE EFFECT

by

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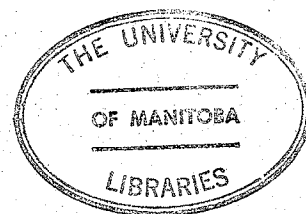


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ABSTRACT

The lifetimes of the $5s5p$, $5s6p$, $5s7p$ and $5s8p$ 1P_1 levels of neutral strontium, and $5p$ $^2P_{3/2}$ and $5p$ $^2P_{1/2}$ levels of the strontium ion have been determined using the Hanle effect. The related oscillator strengths of transitions from these levels have been calculated from the measured lifetimes. The depolarization cross section between the collisions of normal strontium atoms with atoms in the excited $5s5p$ 1P_1 level, and between the excited $5p$ $^2P_{3/2}$ strontium ions with atoms in the excited $5s5p$ 1P_1 level and with atoms in the ground level have also been determined. The results are

$$\begin{aligned}\tau(5s5p \ ^1P_1) &= 4.68 \pm 0.10 \text{ ns} \\ \tau(5s6p \ ^1P_1) &= 3.64 \pm 0.14 \text{ ns} \\ \tau(5s7p \ ^1P_1) &= 4.93 \pm 0.35 \text{ ns} \\ \tau(5s8p \ ^1P_1) &= 5.46 \pm 0.17 \text{ ns} \\ \tau(5p \ ^2P_{3/2}) &= 5.63 \pm 0.17 \text{ ns} \\ \tau(5p \ ^2P_{1/2}) &= 6.74 \pm 0.20 \text{ ns}\end{aligned}$$

$$\begin{aligned}f(4607) &= 1.94 \pm 0.06 \\ f(7169) &= 1.22 \pm 0.05 \\ f(2932) &= (6.60 \pm 0.22)10^{-3} \\ f(5331) &= 0.485 \pm 0.04 \\ f(2570) &= (1.38 \pm 0.04)10^{-2} \\ f(4755) &= 0.34 \pm 0.02 \\ f(2428) &= (4.10 \pm 0.13)10^{-2} \\ f(4077) &= 0.83 \pm 0.03 \\ f(4215) &= 0.37 \pm 0.02\end{aligned}$$

$$\sigma_V(^1S_0 - 5s5p \ ^1P_1) = [(5.2)10^{-7}/\bar{v}] \text{ cm}^2$$

$$\sigma(5p \ ^2P_{3/2} - ^1S_0; 5s5p \ ^1P_1) = 8400 \text{ \AA}^2$$

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CHAPTER I INTRODUCTION

Since the pioneering work of Balmer (1885) on the spectrum of hydrogen, and the demonstration of the existence of series of lines in many spectra by Rydberg (1889), there has been a considerable development in our knowledge of atomic spectra and the associated theory. Measurements of the wavelengths of the spectral lines of many atoms and ions have led to the compilation of energy level tables by Moore (1952). Bohr (1911) using quantum theory provided the first acceptable description of the hydrogen atom. Difficulties with the Bohr theory, particularly with atoms with more than one electron, have led to the rapid development of quantum mechanics. Among the many books on quantum mechanics the one by Condon and Shortley (1936) is still outstanding.

The first observation of the effect of a magnetic field on the radiation emitted by an atom was reported by Zeeman (1897). In 1905 Wood showed that the D lines of sodium could excite D line fluorescence in sodium vapour. Rayleigh (1922) showed that the resonance fluorescence of the 2537 Å line in mercury vapour was polarized, while Wood and Ellet (1923) showed that a small magnetic field would remove the polarization. In 1924 Hanle explained this depolarization of the resonance radiation in terms of the Zeeman effect and we now call this phenomenon the Hanle effect. Nowadays,

the Hanle effect has become one of the most important methods for the measurement of the lifetimes of the excited atomic energy levels. Before we go into more detail, a short review of lifetime and the related oscillator strength measurements will be helpful.

In the past few decades analyses of atomic spectra have succeeded in describing energy levels, fine structure and hyper-fine structure and isotope shift of many atoms and ions. On the other hand, lifetimes of atomic levels and their related oscillator strengths remain of great interest to experimental and theoretical atomic physicists, because there are many unknowns and large discrepancies between the measured and calculated values. Oscillator strengths are widely used in the astrophysics for the study of stellar structure, and for the determination of ion densities and temperatures in plasma physics. More experimental data with greater accuracy is needed.

By definition, the lifetime of an excited atomic state is the average decay time of that state. All possible radiation transitions to lower energy states must be taken into consideration. If N_0 represents the number of atoms in the excited state m at time $t=0$, then the number remaining at time t is given by

$$(1) \quad N_t = N_0 \exp(-t/\tau_m)$$

where τ_m is the mean lifetime of the excited state m under investigation. The mean lifetime, τ_m , is the reciprocal of the sum of the

Einstein A coefficients (Mitchell and Zemansky 1934), so that,

$$(2) \quad \tau_m = 1 / \left(\sum_n A_{mn} \right) \\ = \sum_n g_m c^3 / \left(g_n 8\pi^2 e^2 v_{nm}^2 f_{nm} \right)$$

Here the summation is taken over all possible transitions to the lower state n; g_m and g_n are the statistical weights of the upper and lower levels respectively; f_{nm} is the absorption oscillator strength related to the corresponding transition; v_{nm} is the transition frequency; m_e and e are the rest mass and charge of the electron and c is the speed of light. Classically f_{nm} is equivalent to the number of classical oscillators in one atom in state n. The detailed explanation is given in Chapter II.

The lifetime of an excited level can be calculated from the measured values of the oscillator strength of transitions to lower levels, or can be measured directly. Various experimental methods have been developed and can be classified as 'relative methods' or 'absolute methods' depending upon whether or not the absolute density of atoms needs to be known.

Relative measurement

There are three principal relative methods, the emission method, the absorption method and the hook method. Results of these methods are proportional to the product of the absolute number of the

atoms per unit volume and the oscillator strength. Unless the density is known accurately, only the relative oscillator strengths can be determined with precision.

In the emission method the line intensities in the arc are measured by precise photographic techniques, and a temperature T is assumed for the arc core so that the excited state populations can be determined by Boltzmann's equation. The relative oscillator strengths of many lines in different spectra have been determined by this method. Eberhagen (1955) extended this method to the absolute measurement of oscillator strength by calculating the atom density in the arc from the Saha equation.

In the absorption method the equivalent widths of the absorption lines are measured. Continuous radiation is passed through an atomic vapour and the number of atoms in the ground level is calculated from the measured temperature and vapour pressure data. King (1940) has successfully used this method for relative oscillator strength measurements of many atoms.

The hook method, which was devised by Rozhdestvenskii and developed by Penkin, has been used to determine many relative oscillator strengths. A Jamin-Mach type interferometer, with a King furnace containing the vapour under investigation in one arm, and an accessory tube with a plane-parallel plate inserted in front of the

other arm, is used. The change of light path by the plate and by the gas leads to the formation of 'hooks' on either side of the spectrum lines. By determining the distance between the turning points of the hooks on either side of the spectral line and the length of the column of atoms, the product of the oscillator strength and the atomic concentration is obtained.

The methods described above have the advantage that the relative oscillator strength of a great number of lines can be obtained at the same time. However, the methods of estimating the atomic density may contain a serious error, which affects the absolute oscillator strengths calculated from these data.

Absolute measurement

The absolute methods are the ones which do not require the knowledge of the absolute number of atoms. They are the direct measurement method, the phase shift method, the optical double resonance method and the level crossing method.

The direct measurement method involves the observation of the intensity change along the path of the atomic beam beyond some point where excitation has occurred. The atoms can be excited electrically or optically, or by using the beam-foil technique. From equation (1), the population of the excited levels decays exponentially with the

time after excitation. Therefore, the emitted light intensity decreases exponentially with distance from the excitation region.

This method is not accurate for lifetimes in the nano-second region because these excited atoms can travel only a short distance during their lifetime.

The phase shift technique uses modulated high frequency resonance radiation as an exciting pulse. The period of the exciting pulse is much greater than the lifetime to be measured. The fluorescent radiation has the same pulse shape as the exciting pulse, but lags in phase by an angle due to the finite lifetime of the excited level. By measuring the phase shift angle, the lifetime of the excited level can be determined.

The double resonance method involves exciting an atom with polarized resonance radiation. If the experiment has been arranged so that only one Zeeman state of the excited level is excited, an external rf oscillating magnetic field with the Larmor precession frequency will transfer the atom to another magnetic state. This modifies the polarization and direction of the fluorescent radiation. The lifetime can be calculated from the half width of the double resonance signal. In double resonance experiments the Doppler width of the rf signal is much smaller than the natural line width and can be neglected. But if the rf magnetic field is strong enough to induce many transitions between Zeeman states during the lifetime, power

broadening occurs and must be eliminated. This can be done by measuring the half width at several power levels of the rf oscillating field and extrapolating to zero field. However, for those excited states with lifetime shorter than 10^{-8} second, an rf magnetic field of magnitude about 100 gauss is required to saturate the resonance. The difficulty of the construction and stabilization of such high power rf sources limits the application of the double resonance method to measurements of relatively long lifetimes.

The level crossing method uses somewhat the same technique as the double resonance method, but the need of rf power is eliminated. When two levels of the excited state are tuned to degeneracy by an external magnetic field, interference effects change the spatial distribution of the scattered radiation. By analysing the scattered light signal during a level crossing, the lifetime of an excited state can be obtained accurately.

The most commonly used example of this method is the so called 'Hanle effect' or 'level crossing at zero magnetic field'. The Hanle effect is one of the most powerful methods for precise lifetime measurements. The reasons for this are multifold. First, only very simple equipment is required and no rf broadening, cascade problems and Doppler broadening have to be considered. Second, unlike the relative method, it does not require a direct knowledge of the atom density. Third, it is particularly suitable for lifetimes in the 10^{-8} to 10^{-9}

second range where other methods, also independent of absolute atom density, begin to lose their accuracy. Finally, if the excited level has more than one branch, detecting any one of the branches by the Hanle effect gives the lifetime of the excited level, so that convenient experimental choices can be made.

The history of the Hanle effect goes back to 1921 when Hanle worked a classical theory explaining the influence of a magnetic field on the polarization of resonantly scattered light. The observations were difficult before the photomultiplier tube was invented, and this technique was forgotten until the 1960's. In 1959 Colgrove et al. accidentally found out that the level crossing effect can be explained by the Breit formula which was developed in the 1930's, and with this came a rediscovery of the Hanle effect, which is a special case of level crossing when the levels cross at zero magnetic field. In recent years, many accurate lifetimes for the excited levels of the group I and group II atoms, using the Hanle effect, have been reported. The same technique has also been applied successfully to the group III elements and some diatomic molecules. Experiments for Hanle effect of excited ionic levels have been performed as well. Review articles discussing the use of the Hanle effect have been written by Budick (1967), zu Putlitz (1965), Happer (1968) and Series (1969).

The strontium atom

Atomic strontium has a $5s^2 \ ^1S_0$ ground level. The singlet metastable level $5s4d \ ^1D_2$ is about 2.5 eV above the ground level. Like cadmium and barium, strontium also has doubly excited configurations of $mdnp \ ^1P_1$ levels that are close in energy compared to the $5snp \ ^1P_1$ levels. The mixing of levels of the same parity may produce interesting results in the lifetimes of the 1P_1 levels and the related oscillator strengths. Prior to this work, Eberhagen (1955) studied the oscillator strengths for the $^1P_1 \rightarrow ^1D_2$ transitions by the emission method. Ostrovskii and Penkin (1958, 1961b) determined the relative and absolute oscillator strengths for $5snp \ ^1P_1 \rightarrow ^1S_0$ transitions by hook method. Their absolute values do not agree with the recent Hanle effect or phase shift experiments, but the hook method relative values should be reliable. These results show that the oscillator strengths for the principal series of strontium are not monotonic, and that a minimum value occurs at the second 1P_1 level. Except for the $5s5p \ ^1P_1 \rightarrow ^1S_0$ transition, all the $5snp \ ^1P_1 \rightarrow ^1S_0$ transitions have absorption oscillator strengths close to the forbidden transitions.

De Zafra et al. (1962), Lurio et al. (1964) and Hulpke et al. (1964) have determined the lifetime of the $5s5p \ ^1P_1$ level. Low atomic densities were employed in their experiments, but coherence narrowing, which is eliminated by extrapolation to zero atomic density to obtain true lifetimes, was not studied. Also, the lifetimes of 1P_1

levels higher than 5s5p were not determined. Prompted by this lack of information, the work described here has been undertaken to obtain further information about the lifetimes of the 1P_1 levels of the strontium atom and the related oscillator strengths. First of all, the true lifetime of the first 1P_1 level is obtained from coherence narrowing studies. This gives an accurate absolute oscillator strength for the first $^1P_1 \rightarrow ^1S_0$ transition of wavelength 4607 Å. By coupling this with the accurate relative ratios given by Penkin (1961) from the hook method, the absolute oscillator strengths of the other $^1P_1 \rightarrow ^1S_0$ transitions are calculated. The lifetimes of the 5s6p 1P_1 , 5s7p 1P_1 and 5s8p 1P_1 levels are determined with the same technique, and using equation (2) the upper limit of oscillator strengths of the $^1P_1 \rightarrow ^1D_2$ transitions are determined. As well, the collision broadening cross-section between Sr(5^1P_1) and Sr(1S_0) atoms is studied using the 4607 Å transition.

The levels which are involved in these experiments are shown in Figure 1.

The strontium ion

The singly ionized strontium atom has a $5s^2S_{1/2}$ ground level. The energy levels involved in the present work are shown in Figure 2. The doublet separation between $5p^2P_{1/2}$ and $5p^2P_{3/2}$ is large enough to be resolved easily with our apparatus. Due to the large separation of the 2P levels one may expect that the lifetimes of these two

levels will differ by a few percent. The excitation energy of the $5p \ ^2P_{3/2}$ is 3.04 eV above the ground level. This energy is close to the ionization energy of the neutral strontium atom in the $5s5p \ ^1P_1$ excited state, which is known to be 3.00 eV. This accidental resonance may cause interesting results in the collisions between strontium ions and strontium atoms. The Hanle effect is applied to study these two problems.

Yu. I. Ostrovskii and N. P. Penkin (1961a) have measured the absolute oscillator strengths for the doublet resonance lines of the strontium ion using the hook method. Gallagher (1967a) has measured the lifetimes of the $5p \ ^2P$ levels of the strontium ion and the collision cross section between ions in these levels and argon atoms. However, several extrapolations were made in the analysis of the data, and collisions between strontium ions and strontium atoms were not considered. The present work was undertaken to check the lifetimes and to obtain the information for collisions between strontium ions and strontium atoms utilizing a simpler approach.

Figure 1.

Energy level diagram for the singlet levels of neutral strontium.

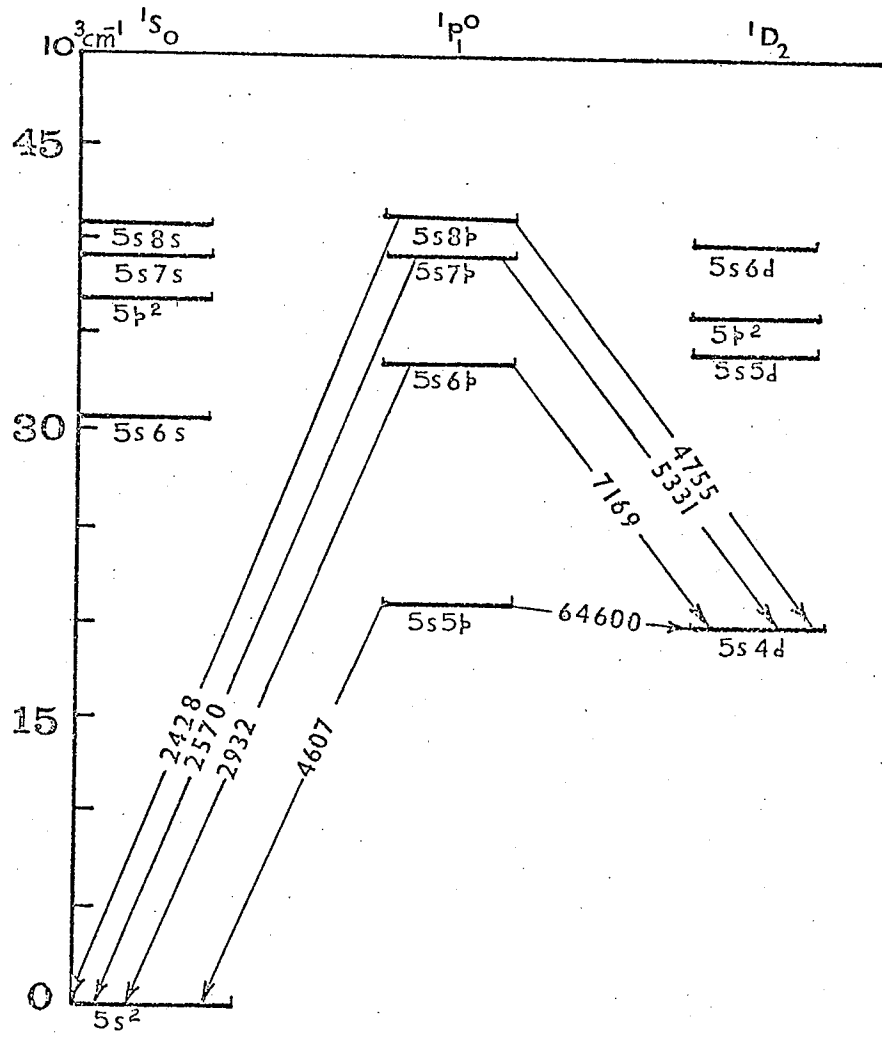
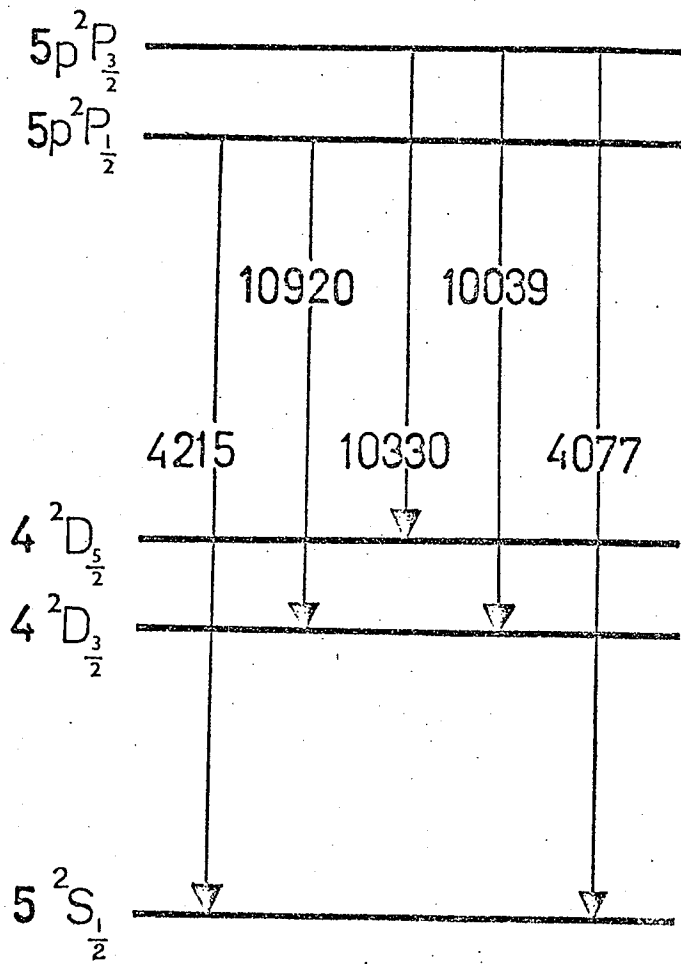


Figure 2.

Partial energy level diagram for the strontium ion.



CHAPTER II THEORY

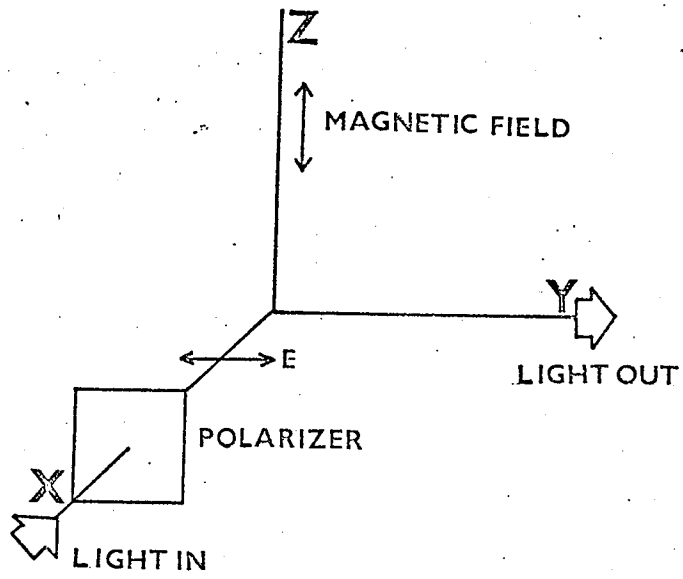
The theory of the Hanle effect based on the classical depolarization concept has been discussed in the book by Mitchell and Zemansky (1934). Quantum mechanically, the Hanle effect can be explained by interference effects, and the theory has been given in a form useful for atomic spectroscopy by Franken (1961) and Lurio et al. (1964).

Hanle effect in the ideal case

The essentials of the classical approach to the Hanle effect are shown in Figure 3. If the incident exciting radiation is polarized with its electric vector along the y axis, and if a sample atom located at the origin is acting as a classical oscillating dipole, after excitation it will vibrate parallel to the direction of polarization of the exciting light. At zero magnetic field this oscillating dipole gradually damps down due to radiation losses, but, the oscillation axis always points in the initial direction. Dipoles do not radiate along the axis of their oscillation, so that nothing will be observed if one looks along the oscillation axis for fluorescent light. However, if there is a magnetic field along the z axis, the dipole excited by the radiation will precess about the field direction at the Larmor precession frequency,

Figure 3.

Geometry of the Hanle experiment.



$\omega_L = \gamma H = g_j \mu_o H / \hbar$, where γ is the gyromagnetic ratio, μ_o is the Bohr magneton and g_j is the Landé g factor for the level. The radiation pattern in the x-y plane is determined by $\sin^2 \theta$, where θ is the angle between the observation direction and the y axis. If the observer stays fixed, θ can be replaced by $\omega_L t$, where t is the time elapsed since excitation started. To obtain the observed intensity for a collection of atoms, a damping term $\exp(-t/\tau)$ must be included where τ is the mean lifetime of the level involved. For continuous excitation and observation, the intensity of the scattered radiation observed along y axis is given by :

$$(3) \quad I = C \int_0^{\infty} \exp(-t/\tau) \sin^2 \omega_L t \cdot dt$$

$$= I_o \left\{ 1 - \frac{1}{1 + (2\gamma\tau H)^2} \right\}$$

which is the inverted Lorentzian distribution. I_o represents the maximum observed intensity of the scattered radiation which depends on the incident light intensity, the beam density, the oscillator strength of the transition involved and also the geometry of the scattering chamber. When H takes the value such that $2\gamma\tau H = 1$, $I = I_o/2$. If we denote this H by $H_{1/2}$, we have

$$(4) \quad 1/\tau = g_j \mu_o \Delta H / \hbar \quad \text{sec}^{-1}$$

where $\Delta H = 2H_{1/2}$ and is the full width at half maximum of the Lorentzian curve. Therefore, in an experiment, one can determine the

lifetime of the excited level from equation (4) by measuring ΔH from the Hanle signal. In many simple atomic spectra the g_j values of excited levels are either well known from theoretical consideration or have been measured by the Zeeman effect or optical double resonance experiments.

Quantum mechanically, the Hanle effect can be described by the Breit-Franken formula (Franken 1961)

$$(5) \quad R \sim \sum_{m, \mu, \mu', m'} \frac{f_{\mu m} f_{m \mu'} g_{\mu' m'} g_{m' \mu}}{1 + i\tau\omega(\mu, \mu')}$$

In the above equation R is the rate at which photons of polarization \hat{f} are absorbed and photons of polarization \hat{g} are emitted in the resonance fluorescence. The subscripts μ and μ' represent the magnetic states of the excited level which can be excited from the state with magnetic quantum number m of the initial level, and which can subsequently decay to the final state with magnetic quantum number m' . If m' does not belong to the same initial level as m , equation (5) is still valid provided that the summation over m is taken only over the magnetic states of the initial level, and the m' summation is over the magnetic states of the final level. The transition frequency $\omega(\mu, \mu') = (E_{\mu} - E_{\mu'})/\hbar$ is between the states μ and μ' . The f 's and g 's are the matrix elements for the absorption and re-emission process in electric dipole transitions; for

example, $f_{\mu m} = \langle \mu | \hat{f} \cdot \vec{r} | m \rangle$ and $g_{\mu' m'} = \langle \mu' | \hat{g} \cdot \vec{r}' | m' \rangle$.

Let us consider the simple $J=0 \rightarrow J=1 \rightarrow J=0$ transitions under the geometrical setting in Figure 3. Here, the final state is the same as the initial state and can be denoted by a. There are only two states corresponding to $m_j = \pm 1$ which can be populated and we will denote them as b and c. Then, $m' = m = a$ and the sum over μ and μ' is taken by considering the two Zeeman states b and c. When the applied field is large enough to completely resolve the magnetic states of the excited level we have $\omega(\mu, \mu') \cdot \tau \gg 1$ and equation (5) reduces to

$$R_1 = R_0 \sim |f_{ab} g_{ba}|^2 + |f_{ac} g_{ca}|^2$$

The rate is just the sum of the individual rates and no interference term is present. When the applied field is near the cross-over point, the excited magnetic states are close to one another and cannot be resolved. Now, $\omega(\mu, \mu') \cdot \tau < 1$ and equation (5) becomes

$$\begin{aligned} R_2 &\sim R_0 + \frac{f_{ba} f_{ac} g_{ca} g_{ab}}{1 + i\tau\omega(b,c)} + \frac{f_{ca} f_{ab} g_{ba} g_{ac}}{1 + i\tau\omega(c,b)} \\ &= R_0 + \frac{A}{1 + i\tau\omega(b,c)} + \frac{A^*}{1 - i\tau\omega(b,c)} \end{aligned}$$

where $A = f_{ba} f_{ac} g_{ca} g_{ab}$ and $A^* = f_{ca} f_{ab} g_{ba} g_{ac}$. The interference effect

occurs in the terms containing A and A*. In the case where the matrix product A is real, A*=A and we have

$$R_2 \sim R_0 + \frac{2A}{1 + \tau^2 \omega^2(b,c)}$$

which is the well known Lorentzian distribution. In the case where the matrix product A is imaginary, A*=-A, and we have

$$R_2 \sim R_0 - \frac{2Ai\omega(b,c)}{1 + \tau^2 \omega^2(b,c)}$$

which is a dispersion shaped distribution. In the case where the matrix product is a complex number, a mixed Lorentzian and dispersion distribution will be obtained. A is real, imaginary or complex depending upon the direction and the polarization of the incoming and outgoing light, and all three cases can be realized experimentally.

If any of the matrix elements in A or A* vanish, there will be no interference and $R_2=R_1$. This means that b and c must be populated coherently in an absorption process and also emit coherently for interference to take place in the cross over region. In other words, if the states are made degenerate by the magnetic field, a single photon can excite both states coherently and a finite phase relation exists between the wave functions representing the excited states. This is analogous to the classical phenomenon of a double slit interference pattern where the same photon can be shared by

both slits. Also, it should be noted that the total absorption cross section for the resonance radiation is independent of whether or not the levels are crossed. The only effect of the crossing is to modify the angular distribution of the re-emitted radiation. Furthermore, the Hanle effect is a crossing at zero field so that for this special case the matrix elements are all field independent.

Lurio et al. have considered the external static magnetic field to be along the z axis and have defined a set of complex unit vectors \hat{e} with components

$$\begin{aligned} e_{\pm} &= e_x \pm i e_y = (\cos\theta \cdot \cos\alpha \pm i \sin\alpha) \exp(\pm i\phi) \\ e_z &= -\sin\theta \cdot \cos\alpha \end{aligned}$$

to allow for arbitrary incident and observing directions. The incident light enters along a direction defined by (θ, ϕ) and with the electric vector \hat{e} making an angle α with respect to $\hat{\theta}$. The scattered light is observed along (θ', ϕ') with its electric vector \hat{e}' making an angle α' with respect to $\hat{\theta}'$ as shown in Figure 4. The general expression for the rate of scattering of resonance radiation can be written as

$$(6) \quad R \sim \sum_{\mu, \mu'} \frac{F_{\mu\mu'} G_{\mu'\mu}}{1 + i\tau\omega(\mu, \mu')}$$

where $F_{\mu\mu'} = \sum_m f_{\mu m} f_{m\mu'}$ and $G_{\mu'\mu} = \sum_{m'} g_{\mu' m'} g_{m'\mu}$ are the excitation and re-radiation matrices respectively. Lurio et al. (1964) have

Figure 4.

Coordinate and polarization system used in the
calculation of the Hanle effect.

