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RESEARCH MEMORANDUM

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RESEARCH MEMORANDUM

STARK EFFECTS IN LINE BROADENING

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RM-1779-AEC

23 May 1956

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STARK EFFECTS IN LINE BROADENING*

INTRODUCTION

In classical impact theories[1], the effect of ions on the width of spectral lines is calculated by supposing that an ion produces a Stark field at the place of the radiating atom, and that the change of this field as the ion moves spreads the radiated frequencies into a small continuum. On the other hand, quantum mechanical calculations based on the customary formulas of the theory of radiation yield line widths which seem to ignore the Stark effect. This was evident in the work of Kivel, Bloom and Margenau[2] where the "universal" effect contained no evidence of second-order Stark contributions but only scattering by Hartree potentials between atom and electron, as was shown by Meyerott and Margenau[3]. First-order Stark effects, called polarization, were present in the results of Ref. 2; they arise from the combination of degenerate states which were included in the calculation.

Higher excited states were ignored, and the impression might arise that their inclusion in the ordinary formalism would automatically yield the quantum analogue of the second-order Stark effect obtained by impact theories. This is not the case. Higher atomic states make contributions to the line width only if they can actually be excited, i.e., if the impinging ions are sufficiently energetic or the temperature is high. The effect is then one of quenching the optically active state rather than of Stark perturbations. The reason for this strange result lies in the fact that radiation theory makes use of what is sometimes called the "sudden approximation:" it assumes none of the upper states to be excited at time t = 0 and then switches on the perturbation. Under these conditions, ordinary perturbation theory likewise falsifies the

* This work was performed by the author as a consultant for The RAND Corporation.

second-order Stark effect.

Part I of this note exhibits the difficulty; Part II shows how it can be removed with the use of wave packets for the perturbing ions.

I

The procedure to be employed is similar to that of Ref. 2, Section IV. We wish to describe a system composed of an atom and one external electron or ion (henceforth we speak of an electron), and states designated by capital letters are states of this combined system. Among them we distinguish the upper radiative state A in which the atom is excited (in state 1) and the electron is in state α, which forms part of a continuum. There are higher atomic states r to which the electron-atom interaction can transfer the system; associated with r are electron states ρ, and the full state, (r,ρ) will be labelled R. Finally, there is the ground state of the atom, 0, which is accompanied by electron states α. The assumption is that it can be reached only by radiation from A, and that during this transition the electron remains in the same state α.

We denote "dark" amplitudes (no photons present) by d, bright ones (one photon of frequency ω_{10} is present) by b. The atom-electron perturbation is C, and $\hbar\omega_{AR} = E_A - E_R$. Furthermore, E_A and E_R are taken to be composed additively of atom and electron energies; the interaction is neglected in ω. Thus $E_A = E_1 + E_\alpha$. One then obtains the following equations for the amplitudes:

$$i\hbar \dot{d}_A e^{-iE_A t} = \sum_l b_{l,0} J_{l,10} e^{-i(E_\alpha + E_0 + \hbar\omega_l)t/\hbar} + \sum_R d_R C_{AR} e^{-iE_R t/\hbar}$$

$$i\hbar \dot{d}_R e^{-iE_R t} = \sum_S d_S C_{RS} e^{-iE_S t/\hbar}$$

$$i\hbar \dot{b}_{l,0} = d_A J_{l,01} e^{-i(E_1 - E_0 - \hbar\omega_l)t/\hbar}$$

The J-term in the first equation gives rise to the natural line width which is not of interest in the present context. Putting it equal to zero has no other effect than to ignore the natural width. In the last equation, $J_{\ell,01}$ is the radiation matrix element for the transition from atomic state 1 to 0 with generation of one photon of frequency ω_ℓ, henceforth simply written as ω. With this understanding the basic equations are

$$i\dot{d}_A = \sum_R d_R C_{AR} e^{i\omega_{AR}t} \tag{1}$$

$$i\dot{d}_R = \sum_S d_S C_{RS} e^{i\omega_{RS}t} \tag{2}$$

$$i\dot{b}_0(\omega) = d_A J_{01} e^{i(\omega_{01} + \omega)t} \tag{3}$$

From Eq. (3) we obtain at once the line shape in emission:

$$\underset{t\to\infty}{\text{line}}\ b_0(\omega) = J_{01}\int d_A e^{i(\omega - \omega_{10})t}\,dt = J(\omega) \tag{4}$$

and the intensity distribution is

$$I(\omega) = \left|J(\omega)\right|^2 \tag{5}$$

It will be noticed that $J(\omega)$ is the Fourier transform of the initial amplitude $d_A e^{-i\omega_{10}t}$. In using Eq. (5) certain averages over directions of polarization must be performed by which $\left|J_{01}\right|^2$ is converted into $\overline{\left|J_{01}\right|}^2$. This is not explicitly indicated and will not concern us in the present investigation.

Let the initial state decay exponentially:

$$d_A = e^{-\gamma t} \tag{6}$$

which means that a single state $A = (1,\alpha)$ was surely present at $t = 0$. Eq. (1)

then states

$$\gamma = \frac{i}{\hbar} \sum_R C_{AR} e^{(i\omega_{AR} + \gamma)t} \tag{7}$$

We now solve (2) by successive approximations. If

$$d_R = d_R^{(0)} + d_R^{(1)} + d_R^{(2)} + \cdots \qquad \text{for } R \neq A$$

then

$$d_R^{(0)} = 0$$

and $$\dot{d}_R^{(1)} = -\frac{i}{\hbar} \left[d_A C_{RA} e^{i\omega_{RA} t} + \sum_{S \neq A} d_S^{(0)} C_{RS} e^{i\omega_{RS} t} \right] .$$

But all $d_S^{(0)}$ occurring in the sum are zero, hence

$$d_R^{(1)} = -\frac{i}{\hbar} C_{RA} \frac{e^{i(\omega_{RA} - \gamma)t} - 1}{i\omega_{RA} - \gamma}$$

In computing $\dot{d}_R^{(2)}$ we retain terms with $d_S^{(1)}$ in $\sum_{S \neq A}$ and find

$$d_R^{(2)} = -\hbar^{-2} \sum_S \frac{C_{RS} C_{SA}}{i\omega_{SA} - \gamma} \left[\frac{e^{(i\omega_{RA} - \gamma)t} - 1}{i\omega_{RA} - \gamma} - \frac{e^{i\omega_{RS} t} - 1}{i\omega_{RS}} \right]$$

Hence, from (7) ,

$$\gamma = -\hbar^{-2} \sum_R |C_{RA}|^2 \frac{e^{(i\omega_{AR} + \gamma)t} - 1}{i\omega_{RA} - \gamma} \tag{8}$$

$$+ i\hbar^{-3} \sum_{RS} \frac{C_{AR} C_{RS} C_{SA}}{i\omega_{SA} - \gamma} \left[\frac{e^{(i\omega_{AR} + \gamma)t}}{i\omega_{RA} - \gamma} + \frac{e^{(i\omega_{AS} + \gamma)t} - 1}{i\omega_{RS}} - \frac{e^{(i\omega_{AR} + \gamma)t} - 1}{i\omega_{RS}} \right]$$

The first line of γ represents the first approximation, the next the second.

Consider the first approximation. Clearly $\sum_R$ contains $\sum_\rho$, the sum over electron states and this can be replaced by an integral.

$$\sum_\rho \longrightarrow \int \frac{V}{(2\pi)^3} \cdot 2\pi k_\rho \, dk_\rho \sin \theta_\rho \, d\theta_\rho$$

provided V = volume, θ_ρ is the angle between the wave number vector $\underset{\sim}{k}_\rho$ and the polar axis. Also,

$$\omega_{AR} = \omega_1 + \omega_\alpha - \omega_r - \omega_\rho \text{ , and } \omega_\rho = \frac{\hbar}{2m} k_\rho^2 .$$

Therefore

$$\sum_\rho \rightarrow \frac{V}{(2\pi)^2} \frac{m}{\hbar} \int \sin\theta d\theta \int k_\rho \, d\omega_\rho$$

and

$$\sum_\rho \left| C_{RA} \right|^2 \frac{e^{(i\omega_{AR} + \gamma)t} - 1}{i\omega_{RA} - \gamma} = \frac{Vm}{4\pi^2\hbar} \int \sin\theta d\theta$$

$$\chi \int_0^\infty \left| C_{RA} \right|^2 k_\rho \frac{e^{i(\omega_{1r} + \omega_\alpha - \omega_\rho)t + \gamma t} - 1}{i(\omega_\rho - \omega_{1r} - \omega_\alpha) - \gamma} \, d\omega_\rho$$

At this point we use the formula

$$\frac{e^{i(\omega_o - \omega)t + \gamma t} - 1}{i(\omega_o - \omega) + \gamma} = \pi \, \delta(\omega, \omega_o)$$

which is formally true for sufficiently small γ and large t. Hence the last integration with respect to ω_ρ vanishes if the only value which contributes to it, namely $\omega_\rho = \omega_{1r} + \omega_\alpha$, is smaller than zero. A finite result occurs when $\omega_\alpha \geq \omega_{r1}$, that is, when the perturbing electron has enough energy to excite the r^{th} state.

In the first approximation, then, γ receives contributions only from matrix elements $\left| C_{RA} \right|^2$ between states R and A with equal energy. The result is very similar to the formula for quenching in Ref. 2 but contains a summation over all accessible atomic states. Explicitly,

$$\gamma^{(1)} = - \frac{Vm}{4\pi\hbar^3} \sum_r k_r \int \sin\theta_r d\theta_r \left| C'_{RA} \right|^2 \qquad (9)$$

where $k_r = \sqrt{\frac{2m}{\hbar}(\omega_\alpha - \omega_{r1})}$

and the prime on C indicates that the states $(r\ \rho)$ and (1α) must have the same energy. Notice that C'_{RA} is a function of θ_r.

We now study the second approximation in Eq. (8). It consists of three terms which will be labelled A, B, and C, so that

$$\gamma^{(2)} = i\hbar^{-3} \sum_{RS} (A + B - C)$$

The sum $\sum_\rho A$ is subject to resonance at the same value k_r, and we find

$$\sum_\rho A = -\frac{Vm}{4\pi\hbar} k_r \int \sin\theta_r d\theta_r \frac{C'_{AR} C'_{RS} C_{SA}}{i\omega_{SA} - \gamma}$$

the primes having the same meaning as before. If γ is neglected against $i\omega_{SA}$, one can define a second-order matrix element

$$D'_{RA} = \sum_S \frac{C'_{RS} C_{SA}}{E_A - E_S} \tag{10}$$

and write

$$i\hbar^{-3} \sum_{RS} A = \frac{Vm}{4\pi\hbar^3} \sum_r k_r \int \sin\theta_r d\theta_r\, C'_{AR} D'_{RA} \tag{11}$$

which looks very much like (9).

When $\sum_{RS} B$ is treated in a similar way, it is seen to add to (11) the complex conjugate to that expression. Up to this point, then

$$\gamma = \frac{V}{4\pi} \frac{m}{\hbar^3} \sum_r k_r \int \sin\theta_r d\theta_r \left[\left| C'_{AR} \right|^2 + 2R(C'_{AR} D'_{RA}) \right] \tag{12}$$

Finally, it can be shown that $\sum_{RS} C$ vanishes for sufficiently small γ as the ratio of Γ/ω_{IR}.

In the second approximation, Eq. (12) is the relevent expression for the

line width, and if it is to be finite the resonances indicated by the primes must occur, since otherwise the matrix elements are to be omitted from the summation over r. The foregoing treatment produces no γ at all when the electrons do not have enough energy to excite the higher atomic states. The reason for this defect lies in the use of an improper approximation for $d_R^{(1)}$ and $d_R^{(2)}$, which implies that these amplitudes are exactly zero at $t = 0$.

II

In classical impact theories, the line width arises from the fact that the perturber moves past the radiating atom. But in the stationary electron state employed in I there is no motion. It is clear, therefore, that a quantum calculation which is to be the analog of an impact theory must operate with initial perturber states that are superpositions of stationary states, i.e., diffusing wave packets. The subsequent analysis will bear this out.

As to the initial state, suppose that the atom has the sharp energy E_1, whereas the electron has a distribution of energies ϵ_α. The initial amplitude d_A will therefore be of the form af_α, and a is time-dependent. For f_α we choose a ''thermal'' wave packet as proposed in a previous paper[2]:

$$f_\alpha = f(k)\,\omega dk = \left(\frac{\hbar^2}{2\pi m K T}\right)^{1/4} \omega^{-1/2}\, e^{-\frac{\hbar^2 k^2}{4mKT}}\, \omega dk \qquad (13)$$

where, aside from obvious symbols, K is the Boltzmann constant, k the wave number and ω the number of free-electron states per unit range in k:

$$\omega = \frac{d}{2\pi}$$

d being the distance in which one electron is encountered. In this analysis, $d = n^{-1/3}$ if n is the number of electrons per cc.

We note that

$$\sum_{\alpha} f_{\alpha}^2 = \int f^2(k)\, \omega dk = 1$$

Our failure to write the initial amplitudes in the form $a_\alpha(t) f_\alpha$ implies that the excited atom will change, as a result of the C-perturbation, in a manner independent of the electron state α. So long as the interaction is weak this is a good approximation.

As to the meaning of Eq. (13) we note that the packet

$$\psi(x) = \int f(k)\, \frac{e^{ikx}}{\sqrt{d}}\, \omega dk \qquad (13')$$

represents a Gaussian function proportional to $\exp\left[-\frac{mKT}{h^2} x^2\right]$. This is not a stationary distribution, but one which diffuses in accordance with the Schrödinger equation for a free particle. Eq. (13') is true at time $t = 0$; at earlier and later times the packet is broader. The accetion of charge at time $t = 0$ and its subsequent diffusion will be seen to correspond in the line-broadening theory to a classical passage of a perturber past an atom.

The diffusion process goes on until, in our model, the half-width of the wave packet is about equal to $\underline{d}$. One need not be precise on this point because the result to be obtained depends on the diffusion length only logarithmically. To compute the diffusion time t we note that a packet whose initial width is $s = \frac{\hbar}{\sqrt{mKT}}$, has a width $\left(s^2 + \frac{\hbar^2}{m^2 s^2} t^2\right)^{1/2}$ at time t. On putting this equal to $\underline{d}$ we find for t the value $\frac{dms}{\hbar} = d\left(\frac{m}{KT}\right)^{1/2}$. Hence

$$\tau = n^{-1/3}(m/KT)^{1/2} \qquad (14)$$

First Order Theory

If $\underline{C}$ has diagonal elements with respect to the atomic states which are large in comparison with off-diagonal terms, Eq. (1) controls the change of d_A.

When written out in detail that equation reads

$$i\hbar\, \dot{a}\, f_\alpha = \sum_\beta a f_\beta\, C_{1\alpha,\alpha\beta}\, e^{i(\omega_\alpha - \omega_\beta)t} \tag{15}$$

The index β sums over electron states, and $\omega_\beta = \frac{\hbar}{2m} k_\beta^2$. The summation on the right affects not only $f_\beta\, e^{-i\omega_\beta t}$, but also $C_{1\alpha,1\beta}$. But since C is presumably a slowly varying function of k_β we replace it by its value at $k_\beta = 0$, where f_β has a maximum. Now

$$\sum_\beta f_\beta e^{-i\omega_\beta t} = \omega^{1/2} \left(\frac{2\pi m K T}{\hbar^2}\right)^{-1/4} \int_{-\infty}^{\infty}\int\int \exp\left[\left(-\frac{\hbar^2}{4mKT} - \frac{i\hbar t}{2m}\right) k^2\right] dk$$

$$= \omega^{1/2}\, (b)^{1/4} (1 + i\sigma t)^{-1/2} \tag{16}$$

with the abbreviations

$$b = \frac{8\pi m K T}{\hbar^2}$$

$$\sigma = \frac{2KT}{\hbar}$$

If Eq. (15) is multiplied by f_α and summed over α, the left side becomes $i\hbar\, \dot{a}$, while the right acquires a factor which is the complex conjugate of (16). Hence

$$i\hbar\, \dot{a} = \omega (b)^{1/2}\, (1 + \sigma^2\, t^2)^{-1/2}\; C_{10,10}\, a$$

$$a = \exp\left\{-i\omega \frac{b^{1/2}}{h}\; C_{10,10} \int_{-\tau}^{t} \frac{dt}{(1 + \sigma^2\, t^2)^{1/2}}\right\}, \quad t \leqslant \tau$$

$$= \exp\left\{-i\omega \frac{b^{1/2}}{\hbar\, \sigma}\; C_{10,10} \times \ln\left(\sqrt{1+\sigma^2 t^2} + \sigma t\right) + \text{const.}\right\} \tag{17}$$

Second-Order Theory

Eq. (2), when written in explicit form, is

$$i\hbar\,\dot{d}_{r\rho} = \sum_{\beta} af_{\beta}\, C_{r\rho,1\beta}\, e^{i(\omega_{r1}+\omega_{\rho}-\omega_{\beta})t}$$

which, by virtue of (16) is

$$\omega^{1/2} b^{1/4} (1+i\sigma t)^{-1/2}\, e^{i(\omega_{r1}+\omega_{\rho})t}\, C_{r\rho,10}\cdot a$$

When this is integrated and the result is put into Eq. (1), we have

$$-\hbar^2 \dot{a} f_{\alpha} = \sum_{r\rho} \omega^{1/2} b^{1/4} C_{r\rho,10} C_{1\alpha,r\rho}\, e^{i(\omega_{1r}+\omega_{\alpha}-\omega_{\rho})t} \times$$

$$\int^{t} \frac{e^{i(\omega_{r1}+\omega_{\rho})t}}{(1+i\sigma t)^{1/2}}\, a(t)dt$$

Again, we multiply by f_{α} and sum over α, and the result is

$$\dot{a} = -\omega b^{1/2} \hbar^{-2} \sum_{r\rho} \left|C_{r\rho,10}\right|^2 \frac{e^{i(\omega_{1r}-\omega_{\rho})t}}{(1-i\sigma t)^{1/2}} \int^{t} \frac{e^{-i(\omega_{1r}-\omega_{\rho})t}}{(1+i\sigma t)^{1/2}}\, a(t)dt$$

A partial integration yields

$$\int^{t} \frac{e^{-i\Lambda\tau}}{(1+i\sigma\tau)^{1/2}}\, a(t)d\tau = \frac{i}{\Lambda} \frac{e^{-i\Lambda t}}{(1+i\sigma t)^{1/2}} a(t) - \frac{1}{2}\frac{\sigma}{\Lambda} \int^{t} \frac{e^{-i\Lambda\tau}}{(1+i\sigma\tau)^{3/2}}\, a(\tau)d\tau$$

The approximation made here lies in neglecting $\dot{a}t$ against a, which limits the analysis to times during which a has not markedly changed from its initial value, 1. If $\sigma \ll |\Lambda| = |\omega_{1r}-\omega_{\rho}|$ only the first term on the right is important, and we find

$$\dot{a} = -i\omega b^{1/2} h^{-2} \sum_{r\rho} \frac{\left|C_{r\rho,10}\right|^2}{\omega_{1r}-\omega_{\rho}} (1+\sigma^2 t^2)^{-1/2}\, a \qquad (18)$$

a result wholly analagous to (17), except that $C_{10,10}$ is now replaced by its second-order equivalent

$$\sum_{rp} \frac{|C_{rp,10}|^2}{\hbar(\omega_{1r}-\omega_p)} \quad .$$

Line Width

In Eqs. (17) and (18) we have recovered the starting point of all classical impact theories. The term in the bracket of (17) represents the phase change at time t. When completed ($t > \tau$) this phase change is

$$\varphi = 2\omega \frac{b^{1/2}}{\sigma\hbar} C_{10,10} \ln\left(\sqrt{1+\sigma^2\tau^2} + \sigma\tau\right)$$

In view of Eq. (14) $\sigma\tau = 2n^{-1/3}(mKT)^{1/2}/\hbar \gg 1$, whence

$$\varphi \doteq \frac{2\omega b^{1/2}}{\sigma\hbar} C_{10,10} \ln\left[4n^{-1/3}\frac{(mKT)^{1/2}}{\hbar}\right] \qquad (19)$$

If τ is small in comparison with the free time between electron impacts (condensations of wave packets) the impacts may be treated as sudden changes of phase, and the following analysis may be employed.

We write (5) in the form

$$I(\omega) \alpha \int_{-\infty}^{\infty} a(t_1) e^{i(\omega-\omega_{10})t_1} dt_1 \int_{-\infty}^{\infty} a^*(t_2) e^{i(\omega-\omega_{10})t_2} dt_2$$

$$a = e^{i\phi(t)}$$

where $\phi(t)$ is the sum of all the phase changes that have occurred at t. Then

$$I(\omega) \alpha \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau} C(\tau)$$

where

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$$C(\tau) = \int_{-\infty}^{\infty} a^*(t)a(t+\tau)dt = \int_{-\infty}^{\infty} e^{i\left[\phi(t+\tau)-\phi(t)\right]} dt$$

and $\omega' = \omega - \omega_{10}$. The evaluation of the correlation function for a single act of emission is of course meaningless; what is intended is an expectation value for an ensemble of radiating atoms. We therefore replace $C(\tau)$ by an expectation value with respect to a random variable ε , assumed to designate the members of the various time series (radiative acts with perturbations) of the ensemble. Thus

$$C(\tau) = \int_{\varepsilon} e^{i\phi(t+\tau,\varepsilon)-i\phi(t,\varepsilon)}\, d\varepsilon \qquad (20)$$

The exponent here is simply the number of impacts occurring between t and $t + \tau$ for a given ε .

Suppose now that the probability of <u>one</u> phase shift φ in a small time interval s is q, and that $Ns = \tau$. Clearly, line $q = 0$, and if s is taken small enough the probability that there shall be <u>no</u> impact in s is $p = 1 - q \doteq 1$. Moreover, if ν is the mean number of impacts per second, $q = \nu s$.

From these considerations one finds, by using Newton's well known formula, that the probability for the occurence of n shifts in the interval τ is

$$\binom{n}{N} p^{N-n} q^{n}$$

whence

$$C(\tau) = \sum_{n} \binom{n}{N} p^{N-n} q^{n} e^{in\varphi} = \sum_{n} \binom{n}{N} p^{N-n} (qe^{i\varphi})^{n}$$

$$= (p + qe^{i\varphi})^{N} = \left[1 - (1 - e^{i\varphi})q\right]^{N} = \left[1 - (1 - e^{i\varphi})\nu s\right]^{\frac{\tau}{s}}$$

As $s \to 0$, then

$$C(\tau) = e^{-\nu\tau(1 - e^{i\varphi})} = e^{-u\tau} \qquad \text{if } \tau > 0 \qquad (21)$$

where $u = \nu(1 - e^{i\varphi})$ (22)

We note from (20) that $C(-\tau) = C^{*}(\tau)$; i.e., $C(\tau) = C^{*}(-\tau)$ for $\tau < 0$.

Hence

$$I(\omega) = \int_{-\infty}^{0} e^{(i\omega' + u^*)\tau} d\tau + \int_{0}^{\infty} e^{(i\omega' - u)\tau} d\tau$$

$$= \int_{0}^{\infty} \left[e^{-(i\omega' + u^*)\tau} + e^{(i\omega' - u)\tau} \right] d\tau$$

$$= \frac{2u_r}{(\omega' - u_i)^2 + u_r^2}$$

Here u_r and u_i are, respectively, the real and imaginary parts of u:

$$u_r = \nu(1 - \cos\varphi)$$

$$u_i = -\nu \sin\varphi$$

Except for our value of φ, these are the results of the usual impact theories. They have been obtained by a number of previous authors who used methods which were perhaps less compact than the present.

The similarity of the phase change φ given by Eq. (19) with the corresponding classical expression is very close. If the potential during a perturber transit across $\underline{d}$ is C,

$$\varphi_{class.} \cong \frac{1}{\hbar} C \frac{d}{v}$$

If in (19) we identify $\sqrt{\frac{KT}{m}}$ with v, $(mKT)^{1/2}/\hbar$ with the De Broglie wave length λ, that equation reads

$$\varphi = \frac{2}{\sqrt{2\pi}} \ln\left(4\frac{d}{\lambda}\right) \frac{Cd}{\hbar v} .$$

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