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## Imprisonment of Resonance Radiation in Solids and Gases

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The description of the self-imprisonment of resonance radiation in gases or solids and of the density distribution of excited species depends upon a transmission coefficient  $T(\rho)$ , the probability that a quantum of emitted radiation will traverse a thickness  $\rho$  of material. This coefficient is shown to be given by the rapidly convergent series  $T(\rho) = \sum_n (-k_0 \rho)^n / n! (n+1)^{1/2}$ , where  $k_0$  is a constant characteristic of the excited species and of the temperature. The above expression is used to give the first accurate discussion of resonance radiation confined between infinite parallel planes, and of radiation inside a sphere over a wide range of  $k_0 \rho$  values.

#### I. INTRODUCTION

Resonance radiation is emitted by an excited atom when it makes a transition directly to its ground state. Such radiation is strongly absorbed by other atoms of the same kind when in their normal states. The absorbing atoms are raised to the same state of excitation as the original atom, and can in turn emit the same quantum of energy in order to return to their ground state. Thus, in a volume of gas, the same quantum of energy may be absorbed and reemitted many times over before it reaches the walls of the container. Under these conditions, the radiation is said to be imprisoned.

In a paper bearing nearly the same title as the present one, Holstein has discussed the imprisonment of resonance radiation in terms of a probability  $T(\rho)$  that a quantum of emitted radiation passes through a thickness  $\rho$  of gas. In Holstein's basic equation for  $T(\rho)$ , the absorption coefficient of the gas,  $k(\nu)$ , is explicitly considered as a function of the frequency, so that an averaging of the monochromatic transmission factor,  $e^{-k(\nu)\rho}$ , over the entire frequency spectrum  $P(\nu)$  of the emitted radiation at a given point must be taken. Then

$$T(\rho) = \int P(\nu)e^{-k(\nu)\rho} d\nu . \qquad (1.1)$$

Of the various forms that the frequency variation can assume, this paper will concern itself only with the Doppler-broadened absorption. In the notation employed by Holstein,  $k(\nu)$  is given for Maxwellian velocity distributions by

$$k(\nu) = k_0 \exp\{-[(\nu - \nu_0)/\nu_0]^2(c/v_0)^2\},$$
 (1.2)

where  $v_0^2 = 2RT/M$ , and  $k_0$  depends on  $v_0$  and on known spectral characteristics of the normal and excited atomic states. It can be shown<sup>1</sup> that

$$pP(\nu) = k(\nu) \tag{1.3}$$

may be used when the shape of the resonance line is Gaussian, as is the case with Doppler broadening or for the luminescence of certain solids emitting at low temperatures. The proportionality constant p is determined by the requirement

$$\int P(\nu) \, d\nu = 1 \quad . \tag{1.4}$$

Let

$$x = [(\nu - \nu_0)/\nu_0](c/\nu_0) . (1.5)$$

Then it will follow that

$$k(x) = pP(v) = k_0 e^{-x^2}$$
, (1.6)

with

$$p = \pi^{1/2} k_0 \,, \tag{1.7}$$

whence

$$T(\rho) = \pi^{-1/2} \int_{-\infty}^{\infty} e^{-x^2} e^{-k_0 \rho e^{-x^2}} dx . \qquad (1.8)$$

The further transformation

$$x = (\ln k_0 \, \rho / v)^{1/2} \tag{1.9}$$

results in3

$$T(\rho) = \frac{1}{\pi^{1/2} k_0 \rho} \int_0^{k_0 \rho} \frac{e^{-y} dy}{(\ln k_0 \rho - \ln y)^{1/2}} . \tag{1.10}$$

Let the probability that a quantum emitted at  $\vec{r}'$  be absorbed in a volume  $d\vec{r}$  at the field point  $\vec{r}$  be given by  $G(\vec{r}', \vec{r})d\vec{r}$ . Let the density of excited atoms be given by  $n(\vec{r})$ . Assume the walls of the container are nonreflecting, and further assume that the time of flight of quanta  $(=1/\gamma)'$  is negligibly small compared to the atomic lifetime  $(=1/\gamma)$ . Then there follows a Boltzmann-type integrodifferential equation<sup>1</sup>:

$$\frac{\partial n(\vec{\mathbf{r}})}{\partial t} = -\gamma n(\vec{\mathbf{r}}) + \gamma \int n(\vec{\mathbf{r}}') G(\vec{\mathbf{r}}', \vec{\mathbf{r}}) d\vec{\mathbf{r}}' \qquad (1.11)$$

and

$$G(\vec{\mathbf{r}}, \vec{\mathbf{r}}') = -\frac{1}{4\pi\rho^2} \frac{\partial T}{\partial \rho} , \qquad (1.12)$$

with  $\rho = |\vec{r}' - \vec{r}|$ . As Holstein remarks, Eq. (1.12) "shows how the whole problem of the space-time variation of the density of excited atoms is referred back to the nature of the transmission coefficient  $T(\rho)$ ." It was the lack of a suitable evaluation of  $T(\rho)$  as given in Eq. (1.10), and hence of  $G(\vec{r}', \vec{r})$ , which strongly limited the utility of Holstein's analysis. In Sec. II, a convenient form for numerical work is derived for  $T(\rho)$ .

#### II. EVALUATION OF $T(\rho)$

The form of Eq. (1.10) shows that  $T(\rho)$  is closely related to the  $\Gamma$  function,

$$\Gamma(p) = \int_0^\infty x^{p-1} e^{-x} dx , \quad p > 0 .$$
 (2.1)

Introduce the new variable  $y = \beta e^{-x/\alpha}$ . Then

$$\Gamma(p) = \frac{\alpha^p}{\beta^\alpha} \int_0^\beta y^{\alpha-1} \left( \ln \frac{\beta}{y} \right)^{b-1} dy . \tag{2.2}$$

In particular, for  $p = \frac{1}{2}$ ,  $\alpha = n + 1$ ,

$$\Gamma(\frac{1}{2}) = \frac{(n+1)^{1/2}}{\beta^{\frac{p+1}{2}}} \int_0^\beta y^n \left(\ln\frac{\beta}{y}\right)^{-1/2} dy , \qquad (2.3)$$

and since  $\Gamma(\frac{1}{2}) = \pi^{1/2}$ , it follows

$$\int_0^\beta \frac{y^n dy}{[\ln(\beta/y)]^{1/2}} = \left(\frac{\pi}{n+1}\right)^{1/2} \beta^{n+1} . \tag{2.4}$$

If  $e^{-y}$  in Eq. (1.10) is expanded in a power series, and if  $\beta = k_0 \rho$ , then

$$T(\rho) = \frac{1}{\pi^{1/2}\beta} \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \int_0^{\beta} \frac{y^n dy}{[\ln(\beta/y)]^{1/2}} . \qquad (2.5)$$

Insertion of Eq. (2.4) into Eq. (2.5) leads to the

final result

$$T(\rho) = \sum_{n=0}^{\infty} \frac{(-\beta)^n}{n! (n+1)^{1/2}} , \quad \beta = k_0 \rho .$$
 (2.6)

#### III. PROCEDURE

If attention is restricted to solutions of Eq. (1.11) of the form

$$n(\vec{\mathbf{r}}, t) = n(\vec{\mathbf{r}}) e^{-\beta t} , \qquad (3.1)$$

where  $\beta$  and  $n(\vec{r})$  satisfy

$$(1 - \beta/\gamma) n(\vec{\mathbf{r}}) = \int G(\vec{\mathbf{r}}, \vec{\mathbf{r}}') n(\vec{\mathbf{r}}') d\vec{\mathbf{r}}', \qquad (3.2)$$

then it can be shown, <sup>1</sup> due to the symmetry of  $G(\vec{r}, \vec{r}') = G(\vec{r}', \vec{r})$ , that the problem can be put into a variational form:

$$\epsilon^{-1} \int n(\vec{\mathbf{r}})^2 d\vec{\mathbf{r}} = \int n(\vec{\mathbf{r}})^2 E(\vec{\mathbf{r}}) d\vec{\mathbf{r}}$$

$$+\frac{1}{2}\int\int [n(\vec{r})-n(\vec{r}')]^2G(\vec{r},\vec{r}')d\vec{r}d\vec{r}'$$
, (3.3)

where

$$E(\vec{\mathbf{r}}) = 1 - \int G(\vec{\mathbf{r}}, \vec{\mathbf{r}}') d\vec{\mathbf{r}}'$$
 (3.4)

and

$$\epsilon = \gamma/\beta$$
 . (3.5)

Then, as Holstein comments,  $^4$  since all  $\beta$  are positive, after a sufficient time the lowest eigenvalue will prevail.  $^5$ 

The linear expansion of  $n(\vec{r})$  over a suitable basis set

$$n(\vec{\mathbf{r}}) = \sum_{i=1}^{\infty} a_i n_i(\vec{\mathbf{r}})$$
 (3.6)

leads as usual to a secular equation:

$$||H_{ij} - \epsilon^{-1}S_{ij}|| = 0 , (3.7)$$

where

$$H_{ij} = \int n_i(\vec{\mathbf{r}}) n_j(\vec{\mathbf{r}}) E(\vec{\mathbf{r}}) d\vec{\mathbf{r}} + \frac{1}{2} \int \int \left[ n_i(\vec{\mathbf{r}}) - n_i(\vec{\mathbf{r}}') \right]$$

$$\times [n_i(\vec{\mathbf{r}}) - n_i(\vec{\mathbf{r}}')] G(\vec{\mathbf{r}}, \vec{\mathbf{r}}') d\vec{\mathbf{r}} d\vec{\mathbf{r}}', \quad (3.8)$$

$$S_{ij} = \int n_i(\vec{\mathbf{r}}) n_j(\vec{\mathbf{r}}) d\vec{\mathbf{r}}. \qquad (3.9)$$

Holstein has solved these equations for the case of the infinite slab and of the infinite cylinder.  $^6$  In both cases, his results are valid only in the limit of large  $k_0 \, \rho$ , and involve many simplifying approximations. The present paper restudies the case of the infinite slab and presents results for the more realistic case of the finite sphere. The case of the finite cylinder will be the subject of a future investigation.

### A. Infinite Slab

Consider a gas confined between two infinite parallel planes a distance L apart. It is also convenient to introduce the parameter  $\kappa = \frac{1}{2} k_0 L$ . Then,

due to symmetry,  $n(\vec{r})$  is a function only of z, and the x and y coordinates can be integrated out immediately. The origin of the z coordinate lies midway between the walls. Equation (3.2) becomes

$$(1 - \epsilon^{-1}) n(z) = \int_{T_{c}/2}^{L/2} n(z') H(\tau) dz', \qquad (3.10)$$

where  $\tau = |z - z'|$ , and

$$H(\tau) = -\frac{\partial \mathcal{E}(\tau)}{\partial \tau} \quad , \tag{3.11}$$

with

$$\mathcal{E}(\tau) = \frac{1}{2} \int_0^{\tau/2} T(\tau/\cos\theta) \, d\cos\theta \,, \tag{3.12}$$

with corresponding forms for Eq. (3.6). The insertion of Eq. (2.6) into Eq. (3.12) leads via elementary integrations to rapidly convergent infinite series<sup>7</sup> at the upper limit of integration, but requires the evaluation of

$$V_0 = \lim_{x \to \infty} \ln x + 2^{1/2} \sum_{n=2}^{\infty} \frac{(-x)^{n-1}}{n!(n+1)(n+1)^{1/2}}$$
 (3.13)

at the lower limit. This limit was obtained numerically, and was taken to be

$$V_0 = 0.672785$$
, (3.14)

which is probably correct to at least five significant figures. A limited number of calculations were performed with  $V_0$  taken as 0.672775 or as 0.672795. The  $\epsilon^{-1}$  were found to vary roughly as  $\kappa^3$  for small

TABLE I. Excitation decay. The infinite slab.

к	€	$\frac{n(\pm 1)}{n(0)}$	Half- width	Half- value
0.1	1.25217ª	0.770b	0.471 <sup>b</sup>	
0.2	1.45414	0.716	0.464	
0.3	$1.64785^{+}$	0.675	0.458	
0.4	1.83959	0.641	0.454	
0.5	2.03178	0.613	0.449	
0.6	2.22555	0.588	0.446	
0.7	2.42150	0.565	0.442	
0.8	$2.61995^{+}$	0.545	0.439	
0.9	2.82108	0.527	0.436	
1.0	3.02496	0.510	0.434	
1.25	3.54680	0.473	0.428	0.977 <sup>b</sup>
2.5	6.39117	0.358	0.409	0.887
5.0	12.92522	0.251	0.392	0.826
10.0	27.91580	0.174	0.381	0.790
15.0	44.39137	0.140	0.376	0.779
20.0	61.81655	0.121	0.374	0.774
25.0	79.93121	0.108	0.373	0.772

<sup>&</sup>lt;sup>a</sup>All calculations based on a four-term expansion which included  $n_0$ ,  $n_1$ ,  $n_2$ ,  $n_6$  for  $\kappa \le 5.0$ , and  $n_0$ ,  $n_1$ ,  $n_5$ ,  $n_6$  for  $\kappa \ge 5.0$ .

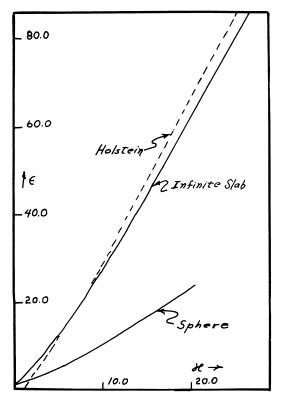


FIG. 1. Display of  $\epsilon$  as a function of  $\kappa$ . Solid lines—this paper; dashed lines—Holstein's approximation [Eq. (4,1)].

changes in  $V_0$ , so that for large  $\kappa$  the results were very sensitive to small changes in the  $V_0$  value adopted. Thus, by interpolation for  $V_0 = 0.672786$ ,  $\epsilon^{-1}$  at  $\kappa = 5.0$  should be 0.00055 higher than the tabular entry.

The expansion basis set is

$$n_0(\xi) = 1$$
 , (3.15)

$$n_i(\xi) = 1 - \xi^{2i}, \quad i \neq 0$$
 (3.16)

where

$$\xi = 2z/L , \qquad (3.17)$$

$$\xi' = 2z'/L$$
 . (3.18)

Note that only  $n_0(\xi)$  contributes to the excited atom density at the walls. All possible four-term combinations of the  $n_i(\xi)$  for i=0 through 6 were employed for values of  $\kappa$  ranging from 0 to 25.0. Double-precision arithmetic was needed for values of  $\kappa$  larger than about 5.0, depending on the choice of basis functions. A limited number of calculations were also performed with three-term expansions, and agreement to five decimals was obtained in the best cases, indicating that the results are probably converged to at least this extent. The results are presented in Table I and Fig. 1, together with other

 $n_6$  for  $\kappa \ge 5$ .0.

<sup>b</sup>All calculations based on a three-term expansion which included  $n_0$ ,  $n_1$ ,  $n_2$  for  $\kappa \le 5$ .0, and  $n_0$ ,  $n_1$ ,  $n_5$  for  $\kappa \ge 5$ .0

data. These other data include  $n(\pm 1)/n(0)$ , the ratio of the density of excited atoms at the surface to the density at the center.

As may be seen from Fig. 1, for large values of  $\kappa$ , the  $\epsilon$  values seem to be becoming linear in  $\kappa$ . For small  $\kappa$  the correct behavior may be closely approximated by the contribution of  $n_0(\xi)$  alone, for which

$$\epsilon^{-1} = H_{00} / S_{00} = 1 - (2^{1/2}/4 + 2V_0) \kappa + 2^{-1/2} \kappa \ln \kappa + O(\kappa^2) ,$$
 (3.19)

$$\epsilon^{-1} = 1 - 0.829284\kappa + 0.707107\kappa \ln \kappa.$$
 (3.20)

For  $\kappa = 0.025$ , Eq. (3.20) gives  $\epsilon = 1.07955$ , which is in a four-figure agreement with the value 1.08002, obtained from the four-term expansion treatment.

#### B. Sphere

Consider a gas contained inside a sphere of radius R. Due to symmetry,  $n(\vec{r})$  is a function only of  $r = |\vec{r}|$ , and the  $\theta$  and  $\phi$  coordinates can be integrated out. Let

$$n(\xi) = \sum a_i n_i(\xi) \tag{3.21}$$

and

$$n_0(\xi) = 1$$
 , (3.22)

$$n_i(\xi) = 1 - \xi^i, \quad i \neq 0$$
 (3.23)

with

$$\xi = \gamma / R . \tag{3.24}$$

Note that i need not be restricted to even values, as was the case with the infinite slab. It seemed more convenient to dispense with the type of coordinate transformation used by Holstein in the study of the infinite slab, and instead to proceed more directly. Let  $\vec{r}$  be the polar axis for  $\vec{r}'$ . Then

$$\rho^2 = r^2 + r'^2 - 2rr'\cos\theta , \qquad (3.25)$$

and Eq. (3.3) can be written as

$$4\pi\epsilon^{-1} \int_0^1 \xi^2 n(\xi)^2 d\xi = 4\pi \int_0^1 \xi^2 n(\xi)^2 d\xi$$
$$+ 2\pi \sum_{n=1}^{\infty} \frac{(-\kappa)^n}{(n-1)!(n+1)^{1/2}} \left(Q_n - \frac{1}{2} P_n\right) , \quad (3.26)$$

where

$$P_{n} = \int_{0}^{1} d\xi \xi^{2} \int_{0}^{1} d\xi' \xi'^{2} [n(\xi) - n(\xi')]^{2} \int_{-1}^{1} d\cos\theta \left(\frac{\rho}{R}\right)^{n-5}$$
(3.27)

and

$$Q_n = \int_0^1 d\xi \, \xi^2 n(\xi)^2 \int_0^1 d\xi' \, \xi'^2 \int_{-1}^1 d\cos\theta \left(\frac{\rho}{R}\right)^{n-3} , (3.28)$$

$$\kappa = k_0 R . ag{3.29}$$

The integrations over  $\cos\theta$  in Eqs. (3.28) and (3.29) lead to terms involving  $|\xi - \xi'|$ , so it is convenient to introduce the variable  $\sigma = s/g$ , where s and g are the smaller and greater of  $\xi$  and  $\xi'$ , respectively. All the integrations can then be expressed in terms of the following auxillary functions:

$$T_i^n = \int_0^1 \sigma^i [(1+\sigma)^n - (1-\sigma)^n] d\sigma$$
, (3.30)

$$L_{i} = \int_{0}^{1} \sigma^{i} \ln \frac{1+\sigma}{1-\sigma} d\sigma , \qquad (3.31)$$

$$N_{ab}^{l} = \int_{0}^{1} \sigma^{l} (1 - \sigma^{a}) (1 - \sigma^{b}) d\sigma , \qquad (3.32)$$

with formally identical functions arising for the integrations over  $\xi$  that remain after the integrations over  $\xi'$  have been performed. For the  $T_l^n$  the following recursion relation is easily established by induction:

$$(n+l+2) T_{t}^{n+1} = 2^{n+1} + (n+1) T_{t}^{n}$$
 (3.33)

The  $L_1$  may be obtained by elementary integrations

$$(2m+1)L_{2m} = 2\ln 2 + \sum_{n=1}^{m} \frac{1}{n},$$
 (3.34)

$$mL_{2m-1} = \sum_{n=1}^{m} \frac{1}{2n-1}$$
 (3.35)

These integrals are most conveniently calculated via

$$(l+1)L_1 = (l-1)L_{1-2} + 2/l$$
, (3.36)

with

$$L_0 = 2 \ln 2, \tag{3.37}$$

$$L_1 = 1$$
 . (3.38)

For Eq. (3.32), write

$$N_{ab}^{l} = \frac{ab(a+b+2l+2)}{(l+1)(a+l+1)(b+l+1)(a+b+l+1)} . \quad (3.39)$$

All possible four-term combinations of the  $n_i(\xi)$  for i=0 through 6 were employed for values of  $\kappa$  ranging from 0 to 20.0. The calculations were performed in both single and double precision as before, the double precision being needed for values of  $\kappa$  larger than about 7.5. The results are presented in Table II and Fig. 1, together with the  $n(\pm 1)/n(0)$  values and other data.

As may be seen from Fig. 1, for  $\kappa$  greater than about 5.0,  $\epsilon$  seems to be becoming linear in  $\kappa$ . For small  $\kappa$  the correct behavior is closely given by the contribution of  $n_0(\xi)$  alone, for which

$$\epsilon^{-1} = H_{00}/S_{00} = 1 - 0.375\sqrt{2}\kappa + O(\kappa^2)$$
, (3.40)

$$\epsilon^{-1} = 1 - 0.530330\kappa$$
 (3.41)

TABLE II. Excitation decay. The sphere.

κ	€	$\frac{n(\pm 1)}{n(0)}$	Half- width	Half- value
0.2	1.11377ª	0.359 <sup>b</sup>	0.727 <sup>b</sup>	0.871 <sup>b</sup>
0.4	1.23334	0.339	0.724	0.855
0.6	1.35856	0.322	0.720	0.841
0.8	1.48910	0.305	0.717	0.829
1.0	1.62475	0.291	0.714	0.818
1.2	1.76524	0.278	0.711	0.808
1.4	1.91035	0.266	0.708	0.800
1.6	2.05983	0.255	0.706	0.792
1.8	2,21349	0.245	0.703	0.785
2.0	2.37110	0.236	0.701	0.778
2.5	2.78123	0.216	0.696	0.765
5.0	5.10424	0.155	0.679	0.727
7.5	7.74700	0.124	0.669	0.711
10.0	10.60647	0.106	0.663	0.702
15.0	16.77703	0.085	0.656	0.692
20.0	23.34635	0.070	0.651	0.688

<sup>a</sup>All calculations based on a four-term expansion which included  $n_0$ ,  $n_2$ ,  $n_5$ ,  $n_6$ .

 $^{\mathrm{b}}$ All calculations based on a three-term expansion which included  $n_0,\ n_2,\ n_5.$ 

For  $\kappa = 0.05$ , Eq. (3.41) gives  $\epsilon = 1.027239$  which is in four-figure agreement with the value 1.027919, obtained from the four-term expansion treatment.

#### IV. DISCUSSION

Holstein's infinite slab results for large  $\rho$  can be written in the present notation as

$$\epsilon = 1.8906 \kappa (\ln \kappa)^{1/2}$$
, (4.1)

so that for  $\kappa=5.0$ , he calculates 11.99. The present results are 12.92. In view of the approximations used by Holstein this is surprisingly good agreement, and his results are presumably more satisfactory for larger  $\kappa$  values. Equation (4.1) is indicated with a dashed line on Fig. 1. The series equation (2.6) for  $T(\rho)$  is numerically convenient for  $\kappa$  values less than about 25. It is shown in the Appendix how the useful range may be doubled, after which results obtained from Holstein's formulas should suffice.

Graphs of the  $n(\vec{r})$  function strongly resemble a Gaussian curve with its tail truncated at the container surface. To help get a feel for the excited atom distribution, half-widths and half-values are entered in Tables I and II. The half-width is the  $\xi_0$  value for which

$$\int_0^{\xi_0} n(\xi) w(\xi) d\xi = \frac{1}{2} \int_0^1 n(\xi) w(\xi) d\xi , \qquad (4.2)$$

where

 $w(\xi) = 1$  for the slab

 $w(\xi) = \xi^2$  for the sphere.

That is, the half-width corresponds to the  $\xi$  value inside which (or outside which) 50% of the excited atoms are to be found. The half-value is the  $\xi_0$  value at which

$$n(\xi_0) = n(0)/2$$
 (4.3)

In a practical experimental situation many factors intervene to make the experimental values differ from the present results. These factors include loss mechanisms in addition to those considered by Holstein, <sup>10</sup> and reflection at the boundaries. In the following paper Menon and Nolle<sup>11</sup> allow for both of these effects with the use of considerations arising from the present calculations. Thus, Menon and Nolle incorporate the effect of additional loss mechanisms into Eq. (1.11) by changing the coefficient of  $n(\vec{r})$  slightly, a modification whose consequence is to merely raise all the  $\epsilon^{-1}$  by a constant amount. They consider the effect of boundary reflection via the surface densities of excited atoms determined here.

A third difficulty in comparison is due to the use of block or cylindrical samples, rather than infinite slabs or spheres. For a cube of side length 2s, a sphere of radius 1.2407s, the radius of a sphere of equal volume, should be a good approximation. For the cylindrical sample, if the ratio of length to face diameter  $(\eta)$  is near 1, then a sphere of radius  $(3\eta/2)^{1/2}R$  (the equivolume sphere) should be a good approximation. However, for wide deviations of  $\eta$  from 1, it would be preferable to have results directly for the cylindrical geometry.

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APPENDIX: SUM RULE AND DUPLICATION FORMULA FOR THE  $T(\rho)$ 

Define

$$\pi^{1/2} T_{\mu}(\beta) = \int_{0}^{1} \frac{z^{\mu - 1} e^{-\beta z} dz}{\left[\ln(1/z)\right]^{1/2}}$$
 (A1)

so that for  $\mu = 1$  and with  $\beta = k_0 \rho$ ,  $T_1(\beta)$  is exactly the transmission coefficient  $T(\rho)$ . Then

$$\sum_{\mu=m}^{\infty} \frac{\pi^{1/2} T_{\mu}(\beta) \beta_{0}^{\mu-m}}{\mu-m!} = \int_{0}^{1} \frac{e^{-\beta z} z^{m-1}}{\left[\ln(1/z)\right]^{1/2}} \sum_{\mu=m}^{\infty} \frac{(\beta_{0} z)^{\mu-m}}{\mu-m!} dz$$
(A2)

$$= \int_{0}^{1} \frac{e^{-(\beta_{0}-\beta)z}z^{m-1}}{[\ln(1/z)]^{1/2}} dz .$$
 (A3)

Hence,

$$T_{m}(\beta - \beta_{0}) = \sum_{\mu = m}^{\infty} \frac{\beta_{\mu}^{0 - m}}{\mu - m!} T_{\mu}(\beta) , \qquad (A4)$$

$$= \sum_{\mu=0}^{\infty} \frac{\beta_0^{\mu}}{\mu!} T_{\mu+m}(\beta) . \tag{A5}$$

$$T_{\mu}(2\beta) = \sum_{l=0}^{\infty} \frac{(-\beta)^{l}}{l!} T_{\mu+l}(\beta) , \qquad (A6)$$

In particular if  $\beta_0$  is set equal to  $-\beta$ , we obtain a duplication formula

and if  $\beta_0$  is set equal to  $\beta$ , we obtain a sum rule

$$\sum_{\mu=1}^{\infty} \frac{\beta^{\mu-1}}{(\mu-1)!} T_{\mu}(\beta) = 1 . \tag{A7}$$

<sup>1</sup>T. Holstein, Phys. Rev. <u>72</u>, 1212 (1947).

 $^{2}$ T. Holstein, Phys. Rev.  $\overline{72}$ , 1212 (1947), Eq. (2.3).

 $^{3}$ T. Holstein, Phys. Rev.  $\overline{22}$ , 1212 (1947), Eq. (2.21).

<sup>4</sup>T. Holstein, Phys. Rev. <u>72</u>, 1218 (1947).

 $^5$ The eigenvalues seem to be well separated. For a typical case ( $\kappa$  = 2.5), reported in the sphere calculation, the second root was nearly double the value of the lowest. The situation becomes more favorable as the radius becomes larger.

<sup>6</sup>T. Holstein, Phys. Rev. 83, 1159 (1961).

<sup>7</sup>The details of the arithmetic manipulation are similar

to the procedures used in the sphere, discussed in text.  ${}^{8}I$  oss of significant figures was encountered due to the near-linear dependence of the higher-order  $N_{I}(\xi)$ . It probably would be more desirable to have chosen the  $N_{I}(\xi)$  to have been the Legendre polynomials of even

<sup>9</sup>T. Holstein, Phys. Rev. <u>72</u>, 1212 (1947), Eq. (4.22).

<sup>10</sup>T. Holstein, Phys. Rev. <u>72</u>, 1212 (1947), Eq. (3.2).

<sup>11</sup>N. S. K. Menon and A. W. Nolle, the following paper, Phys. Rev. B <u>4</u>, 3890 (1971).

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# Radiation Imprisonment and Magnetic Field Effects on Luminescence in Pink Ruby\*

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The theory of imprisoned radiation developed by Holstein and recently extended by Scherr is adapted to apply to the luminescence decay in a solid. In addition to including a correction for losses from the excited level, it is necessary to consider reflections at the sample surfaces. Reflections are calculated approximately for a slab. The single Gaussian absorption function used in the reabsorption theory in lieu of an actual absorption function for the imprisoned radiation has the same integrated absorption I as the actual function and has a height equal to  $\sqrt{2}I$ times the integral of the square of the actual function. Calculations are described for the ruby R lines, including the case where the Zeeman components are separated in a magnetic field. Experimental measurements of decay times at 77 °K of the ruby R lines are presented for samples having a Cr<sup>+3</sup> concentration of approximately 10<sup>19</sup> cm<sup>-3</sup> and thickness from 0.6 to 13 mm. Magnetic fields up to 20 kG are used. The various decay times, lying between 5 and 12 msec, are predicted within about 5% by the theory, with the help of the approximate absorption corrections and of Zeeman component intensities given by Sugano and Tanabe. The  $R_1$  line is used for most comparisons. Intensity increases of up to 100% in the steady-state R-line emission in a 20-kG magnetic field are shown to be accounted for by approximate treatments related to the decay-time theory. No change of transition probabilities with magnetic field is indicated by the foregoing results. Further observations suggest that the channels leading from the excitation levels for both single ions and pairs are affected by magnetic field, however. These observations include changes in the relative intensities of various pair lines when the field is applied, and a field-dependent difference in the emission intensities produced with blue and green excitation. Field-induced changes in pumping light absorption, if present, are insufficient to account for these variations.

#### I. INTRODUCTION

The theory of reabsorption of resonance radiation, also known as imprisonment, has been extensively treated by Holstein. <sup>1</sup> He provided working formulas for the limiting case of nearly complete reabsorption, which is important for gases. The calculations are troublesome for the case of a species having small peak absorption or present at small concentration (cases often encountered with solids).

with the result that empirical approximations have been proposed<sup>2</sup> in lieu of the theory. Recently Scherr<sup>3</sup> has put the Holstein transmission function for a Gaussian line into a form convenient for partial reabsorption. From this he has evaluated the decay-time function for an infinite slab and also has extended the theory and the calculations to the case of a sphere.

We shall show that where sufficient information is available as to line shape and absorption cross