

Alignment of excited atoms in a gas discharge

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Trapping of resonant radiation in a gas discharge leads to alignment of excited atoms. A general expression is derived for the degree of various types of alignment. It is shown that alignment enhances the effective lifetime of the excited atoms. This should become manifest when alignment is destroyed by a weak magnetic field.

1. As is well known, for excited atoms to become oriented and aligned in a gas it is necessary to use excitation with polarized light or at least to have an anisotropic distribution of the directions of the exciting light beams. In the case of excitation with electron impact, an anisotropic electron-velocity distribution is necessary. In the positive column of a gas discharge, excitation of the atoms is by electrons, and their velocity distribution at not too low pressures is practically isotropic. Nonetheless, effects connected with alignment of the excited atoms in a discharge in neon and in a mixture of neon with helium at pressures of approximately 1 mm Hg have recently been observed^[1-3]. A manifestation of alignment is the resonant variation of the intensity of the radiation in weak magnetic fields. Chaika^[4] has proposed to explain the experimental results by means of two possible mechanisms of alignment. Both mechanisms are connected with reabsorption of the photons in the discharge. First, the photon distribution in the discharge tube is, generally speaking, anisotropic. The radiation at the wall is directed mainly perpendicular to the tube axis (from the axis to the wall). At the tube axis, the predominant direction is the direction of the axis. The excited atoms produced upon absorption of such radiation are aligned. The second possible cause of the observed effects is the finite width of the spectrum of the exciting photons. Because of this, each group of atoms with a given velocity direction turns out to be aligned, even if there is no integral alignment for all the atoms. Chaika called this new type of alignment latent alignment. In the case of dragging of the resonant radiation, both types of alignment are due to the fact that the volume is finite.

At a large value of the radiating volume, the photons will be anisotropic in direction only in a relatively small region near the wall, where the concentration of the excited atoms is small. The spectral distribution of photons in the greater part of the volume is much broader than the Doppler absorption contour, so that the latent alignment will also be small.

In the present paper we study the degree of alignment of atoms in an excited state when the resonant radiation is dragged in a finite volume. It is shown that the alignment influences the excited-atom effective lifetime due to the dragging. This should be manifest in a shortening of the lifetime when the alignment is destroyed by a weak magnetic field. According to Holstein^[5], the effective lifetime τ is of the order of

$$\tau \sim k_0 R \tau_0 (\pi \ln k_0 R)^{1/2},$$

where τ_0 is the natural lifetime, R is the dimension of the vessel, and k_0 is the coefficient of absorption at the center of the Doppler contour (it is assumed that $k_0 R \gg 1$). As shown later on, owing to alignment, this time lengthens by an amount $\Delta\tau$, with

$$\Delta\tau / \tau \sim 1 / \ln k_0 R.$$

The effective lifetime should change by an amount of this order if the alignment is destroyed by a weak magnetic field, such that the Zeeman splitting of the excited state becomes of the order of the level width.

2. We start with the equation for the density matrix of the excited atoms $f_{mm'}$, which depends on the coordinates and on the velocity

$$df_{mm'} / dt = -\gamma f_{mm'}(r, v) + \gamma (\hat{\mathcal{L}}\hat{f})_{mm'}. \quad (1)$$

Here the indices m and m' number the Zeeman sub-levels of the considered excited state with angular momentum j_1 , and γ is the reciprocal lifetime of this state. The term containing $(\hat{\mathcal{L}}\hat{f})$ takes into account the dragging of the radiation^[6]:

$$(\hat{\mathcal{L}}\hat{f})_{mm'} = \int d^3v' \int d^3r' \sum_{\substack{m'' \\ m, m_1}} K_{mm''}^{m_1 m_1'}(r - r', v, v') f_{m''m_1'}(r', v'). \quad (2)$$

The kernel K characterizes the probability that an atom having a velocity v will be excited at the point r by a photon emitted by an atom situated at the point r' and having a velocity v' .

The eigenfunctions $\hat{\phi}_N(v)$ and the eigenvalues of the operator $\hat{\mathcal{L}}$ were obtained in^[7] for an unbounded volume:

$$\hat{\phi}_N(v) = \sum_{\kappa L} C_{\kappa L}^{(N)} \Phi_{\kappa L}(v/v_0) \hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi), \quad (3)$$

where v , ϑ , and φ are the spherical coordinates of the velocity, $v_0 = (2kT/M)^{1/2}$, $C_{\kappa L}^{(N)}$ are certain coefficients, the equations for which were derived in^[7], $\Phi_{\kappa L}^{(L)}(v/v_0)$ are functions containing the Laguerre polynomials $L_{(n-L)/2}^{L+1/2}(v^2/v_0^2)$. An explicit expression for these functions is also given in^[7]; $\hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi)$ is a matrix with the indices m and m' :

$$\hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi) = \sum_{qM} (\kappa L JS | \kappa L q M) \hat{T}_q^* Y_{LM}(\vartheta, \varphi).$$

Here $Y_{LM}(\vartheta, \varphi)$ is a spherical function and \hat{T}_q^K is an irreducible tensor operator. The quantity in the parentheses is the Clebsch-Gordan coefficient corresponding to the addition of the angular momenta κ and L . By N we denote the set of quantum numbers characterizing the eigenfunction (n, J, S , parity κ , etc.).

It is convenient to seek the solution of (1) for a finite volume in the form of an expansion in the functions $\hat{\phi}_N(v)$:

$$\hat{f}(r, v) = \sum_N a_N(r) \hat{\phi}_N(v). \quad (4)$$

The density matrix of the entire ensemble of atoms can be obtained by integrating $\hat{f}(r, v)$ with respect to the velocities, in which case a nonzero result is given only

by the functions with $n=0$. For these functions, the sum (3) contains only one term with $L=0$ and $\kappa=J$. The number J characterizes the polarization of the atoms ($J=0$ —population, $J=1$ —orientation, $J=2$ —alignment). The coefficients of these functions will be designated a_0 , a_{1S} , and a_{2S} . For $j_1=1$, the coefficients a_0 and a_{20} are connected with the density matrix $f_{mm'}$ integrated over the velocities in the following manner:

$$a_0 = \frac{1}{\pi^{3/2} \gamma_3} (f_{11} + f_{-1-1} + f_{00}), \quad (5)$$

$$a_{20} = \frac{1}{\pi^{3/2} \gamma_6} (f_{11} + f_{-1-1} - 2f_{00}).$$

All the remaining functions for which $n \neq 0$ make no contribution to the polarization moments of the entire ensemble. It can be stated that these functions correspond to "latent" polarization moments. Using (1) and (5), we obtain an equation for the coefficients $a_N(\mathbf{r})$:

$$\frac{da_N}{dt} = -\gamma a_N(\mathbf{r}) + \gamma \sum_{N'} \hat{K}_{NN'} a_{N'}, \quad (6)$$

where

$$\hat{K}_{NN'} a_{N'} = \int d^3r' K_{NN'}(\mathbf{r}-\mathbf{r}') a_{N'}(\mathbf{r}'), \quad (7)$$

$$K_{NN'}(\mathbf{r}) = \int d^3v \int d^3v' \sum_{mm'm_1m_1'} [\hat{\varphi}_N(\mathbf{v})]_{mm'} K_{mm'}^{(N)}(\mathbf{r}, \mathbf{v}, \mathbf{v}') [\hat{\varphi}_{N'}(\mathbf{v}')]_{m_1m_1'}. \quad (8)$$

The calculation of $K_{NN'}(\mathbf{r})$ with the aid of formula (20) of [7] leads to the result

$$K_{NN'}(\mathbf{r}) = \frac{\gamma(2J+1)(2J'+1)}{8\pi} \sum_{q\nu\nu'} x_{q\nu\nu'}^{(N)} x_{q\nu\nu'}^{(N')} D_{q,-q}^J(\mathbf{n}) D_{q,-q}^{J'}(\mathbf{n}) I_{nn'}(|\mathbf{r}|). \quad (9)$$

Here $D_{Sg}^J(\mathbf{n})$ is the matrix of rotation from the initial system of coordinates to a system in which the z axis is directed along the vector \mathbf{r} ; \mathbf{n} is a unit vector along \mathbf{r} . The function $I_{nn'}(|\mathbf{r}|)$ is determined by the expression

$$I_{nn'}(|\mathbf{r}|) = \frac{1}{l_0 |\mathbf{r}|^2} \int_{-\infty}^{\infty} dx \exp\{-\mathcal{F}(x)|\mathbf{r}| - 2x^2\} \tilde{H}_n(x) \tilde{H}_{n'}(x), \quad (10)$$

where l_0 is the mean free path (the reciprocal of the absorption coefficient) of the photon at the line center, $\mathcal{F}(x) = (1/l_0) \exp(-x^2)$, and $\tilde{H}_n(x)$ is a normalized Hermite polynomial. The numbers $x_{q\nu\nu'}^{(N)}$ are connected with the coefficients $C_{KL}^{(N)}$ by the relation

$$x_{q\nu\nu'}^{(N)} = \sum_{\kappa L} (-1)^{L-\kappa} B_{\kappa L}^{q\nu\nu'}(J) b_n^L C_{\kappa L}^{(N)}, \quad (11)$$

where

$$b_n^L = (-1)^{(n-L)/2} \pi^{1/2} \left[n! / 2^{n+1} \left(\frac{n-L}{2} \right)! \Gamma \left(\frac{n+L+3}{2} \right) \right]^{1/2}, \quad (12)$$

$$B_{\kappa L}^{q\nu\nu'}(J) = 3[(2j_1+1)(2\kappa+1)(2L+1)]^{1/2} \begin{Bmatrix} \kappa & 1 & 1 \\ j_0 & j_1 & j_1 \end{Bmatrix} \times \begin{pmatrix} \kappa & 1 & 1 \\ -q & -\nu & \nu' \end{pmatrix} \begin{pmatrix} \kappa & L & J \\ -q & 0 & q \end{pmatrix}. \quad (13)$$

From (23) of [7] we can obtain the following equation for $x_{q\nu\nu'}^{(N)}$:

$$\frac{1}{2} \sum_{q\nu\nu'} R_{q\nu\nu'}^{q\nu\nu'} x_{q\nu\nu'}^{(N)} = (1-\lambda_N) x_{q\nu\nu'}^{(N)}, \quad (14)$$

where

$$R_{q\nu\nu'}^{q\nu\nu'} = \sum_{\kappa L} B_{\kappa L}^{q\nu\nu'}(J) B_{\kappa L}^{q\nu\nu'}(J) (b_n^L)^2, \quad (15)$$

λ_N is the eigenvalue corresponding to the function $\hat{\varphi}_N(\mathbf{v})$. The normalization condition for the quantities x is

$$\frac{1}{2} \sum_{q\nu\nu'} [x_{q\nu\nu'}^{(N)}]^2 = 1 - \lambda_N. \quad (16)$$

The indices ν and ν' run here through the values ± 1 , and the index q takes on the values 0 and ± 2 .

It follows from (10) that the dragging operator interrelates only states having the same parity of the number n . From (11) and (13) we see the following symmetry properties of the numbers x :

$$x_{011}^{(N)} = (-1)^{L+J} x_{0-1-1}^{(N)}, \quad x_{2-1-1}^{(N)} = (-1)^{L+J} x_{211-1}^{(N)} \quad (17)$$

(we recall that the parity of L is a quantum number). These relations enable us to reduce the system (14) to two equations with two unknowns.

In the case of strong dragging (the only one considered in the present paper), when $k_0 R \gg 1$, all the off-diagonal matrix elements $K_{NN'}$ are small, since as $R \rightarrow \infty$ we have

$$\int d^3r' K_{NN'}(\mathbf{r}-\mathbf{r}') = (1-\lambda_N) \delta_{NN'}$$

in accordance with the results of [7]. Thus, at $k_0 R \gg 1$ we can replace (6) by the following approximate system of equations:

$$\frac{da_0}{dt} = -\gamma(1-\hat{K}_{00})a_0 + \gamma \sum_{N \neq 0} \hat{K}_{N0} a_N, \quad (18)$$

$$\frac{da_N}{dt} = -\gamma(1-\hat{K}_{NN})a_N + \gamma \hat{K}_{N0} a_0. \quad (19)$$

From (9) follows a simple expression for $K_{0N}(\mathbf{r})$:

$$K_{0N}(\mathbf{r}) = K_{N0}(\mathbf{r}) = x_{011}^{(N)} Y_{J_S}(\mathbf{n}) \frac{1}{2\pi^{3/4}} I_{n0}(|\mathbf{r}|), \quad (20)$$

where $Y_{J_S}(\mathbf{n})$ is a spherical function that depends on the direction of the vector \mathbf{r} . Formula (20) was obtained with allowance for relations (17) and is valid only for even values of $L+J$, for otherwise $K_{0N}(\mathbf{r}) \equiv 0$. It follows from (13) that x_{011} is nonzero only at even values of the sum $\kappa+L+J$. Thus, $K_{0N}(\mathbf{r})$ differs from zero only for those sets of the quantum numbers N for which the numbers n , κ , L and J are even. In the case of strong dragging we can replace $\hat{K}_{NN'} a_{N'}$ in (18) by $(1-\lambda_N) a_N$, where λ_N is the eigenvalue obtained in [7]. All the λ_N at $N \neq 0$ are of the order of unity, and therefore we can neglect the term da_N/dt in (19), since the interesting characteristic time of variation of the population is much larger in the case of strong dragging than the natural lifetime γ^{-1} . Then

$$a_N = \lambda_N^{-1} \hat{K}_{N0} a_0. \quad (21)$$

Calculation of the function $\hat{K}_{N0} a_0$ in the case of strong dragging is best carried out in the following manner: using formulas (20) and (10), and reversing the order of integration, we obtain

$$\hat{K}_{N0} a_0 = \frac{x_{011}^{(N)}}{2\pi^{3/4}} \int_{-\infty}^{\infty} dx e^{-x^2} \tilde{H}_n(x) \int d^3r' a_0(\mathbf{r}') \frac{e^{-\mathcal{F}(x)|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|^2} \mathcal{F}(x) Y_{J_S}(\mathbf{n}). \quad (22)$$

Here V is the volume of the vessel (we recall that \mathbf{n} is the unit vector along the direction of $\mathbf{r}-\mathbf{r}'$). We shall henceforth take the unit length to be the characteristic vessel dimension R , and introduce the parameter

$$k_0 R = R/l_0 = \alpha \gg 1. \quad (23)$$

We make in (22) the change of variables $\alpha \exp(-x^2) = t$; then

$$\hat{K}_{N0} a_0 = \frac{x_{011}^{(N)}}{2\pi^{3/4}} \frac{1}{\alpha} \int_0^\alpha dt \frac{\tilde{H}_n(\sqrt{\ln(\alpha/t)})}{\sqrt{\ln(\alpha/t)}} \int d^3r' a_0(\mathbf{r}') \frac{t e^{-t|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|^2} Y_{J_S}(\mathbf{n}). \quad (24)$$

At large α , it would be natural to extend the integral

with respect to t to infinity. But the integral with respect to the coordinates in (24) does not tend to zero as $t \rightarrow \infty$. To get around this difficulty, we break up $a_0(\mathbf{r}')$ into two terms:

$$a_0(\mathbf{r}') = [a_0(\mathbf{r})] + [a_0(\mathbf{r}') - a_0(\mathbf{r})]. \quad (25)$$

In that part of (24) which contains the first term, the integral with respect to the coordinates \mathbf{r}' can be replaced by an integral over the region that is external with respect to the vessel, taken with the minus sign. Indeed, the integral over all of space vanishes if the numbers n and J do not vanish simultaneously.

We can now extend the integral with respect to t to infinity and neglect $\ln t$ in comparison with $\ln \alpha$. We then obtain

$$a_N(\mathbf{r}) = \frac{1}{\lambda_N} \frac{x_{011}^{(N)}}{\pi^n} \frac{\mathcal{H}_n(\sqrt{\ln \alpha})}{\alpha \sqrt{\ln \alpha}} \Psi_{JS}(\mathbf{r}), \quad (26)$$

where

$$\Psi_{JS}(\mathbf{r}) = -\frac{1}{\sqrt{4\pi}} a_0(\mathbf{r}) \int_{V'} d^3\mathbf{r}' \frac{Y_{JS}'(\mathbf{n})}{|\mathbf{r} - \mathbf{r}'|^4} + \frac{1}{\sqrt{4\pi}} \int_V d^3\mathbf{r}' [a_0(\mathbf{r}') - a_0(\mathbf{r})] \frac{Y_{JS}'(\mathbf{n})}{|\mathbf{r} - \mathbf{r}'|^4}. \quad (27)$$

The second integral is taken here over the volume of the vessel V , and the first over the region V' which is external with respect to the vessel. The second integral contains a logarithmic divergence at $\mathbf{r} = \mathbf{r}'$, due to the first term of the expansion of $[a_0(\mathbf{r}') - a_0(\mathbf{r})]$ in powers of $\mathbf{r} - \mathbf{r}'$. This divergence is eliminated by taking the integral in the sense of the principal value, i.e., by separating the region that is symmetrical with respect to the point \mathbf{r} , and then letting the dimension of this region approach zero. At even J , this region makes a zero contribution to the integral with respect to the coordinates of formula (24) at arbitrarily large t .

We recall that in formulas (26) and (27) the quantities \mathbf{r} and \mathbf{r}' are measured in units of R .

Formula (26) enables us to estimate the order of magnitude of a_N . We see that a_N behaves with increasing α like $\alpha^{-1}(\sqrt{\ln \alpha})^{n-1}$. In the calculation of Ψ_{JS} we used an expression given by Holstein^[5] for the concentration of the excited atoms. For a flat layer of thickness $2R$ we then have $a_0 = c(1 - \xi^2)$, where $\xi = z/R$ and c is a constant.

Calculation by means of formula (27) yields

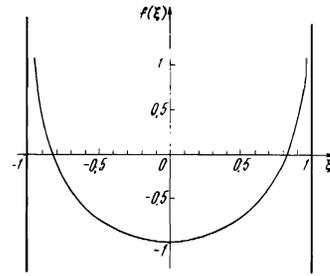
$$\Psi_{JS}(z) = \delta_{s,0}(-1)^{J/2} \frac{\sqrt{2J+1}(J+1)!!}{(J-1)!(J+2)!!} \left[2 + \xi \ln \frac{1-\xi}{1+\xi} \right] c. \quad (28)$$

The quantization axis is perpendicular here to the plane of the layer. The divergence in this expression on the boundaries of the vessel is connected with the fact that the transformations used in the derivation of formula (26) are not valid near the boundary, in a layer of thickness on the order of several l_0 .

The alignment that is integral with respect to the velocities is determined by the coefficient a_N at $n=0$ and $J=2$. We then have in formula (26) $x_{011} = 1/\sqrt{10}$ and $\lambda = 0.3$ for the transition $j_1 = 1 \rightarrow j_0 = 0$. The integral alignment depends on the dimensions of the vessel and on the absorption coefficient k_0 like $(k_0 R \sqrt{\ln k_0 R})^{-1}$ if $k_0 R \gg 1$. In the central plane of the layer (the alignment axis is perpendicular to the plane of the layer) we have

$$a_{20}(0) = -c[1, 2\alpha(2\pi \ln \alpha)^{3/2}]^{-1}. \quad (29)$$

The dependence of the alignment on z (the distance from the central plane of the layer) is shown in the figure.



Dependence of the degree of alignment in a flat layer of thickness $2R$ on the coordinate $\xi = z/R$, $f(\xi) = -a_N(z)/a_N(0)$.

This dependence can be interpreted easily when the angular momentum of the excited state is $j_1 = 1$. Then a_{20} is connected by relation (5) with the density matrix integrated over the velocities. It is seen from the figure that near the central plane of the layer, the predominantly populated sublevel is the one with $m=0$, and at the layer boundary, to the contrary, population of the sublevels $m = \pm 1$ predominates. When integrated over the layer, the population of the level $m=0$ predominates. The distribution of the alignment in a cylinder has a complicated character^[4]. Calculations with the aid of formulas (26) and (27) yield the following value for the alignment on the axis of an infinite cylinder (in the limit of strong dragging) for the transition $j_1 = 1 \rightarrow j_0 = 0$:

$$a_{20}(0) = \pi c [4, 8\alpha(2\pi \ln \alpha)^{3/2}]^{-1}. \quad (30)$$

The alignment axis is in this case directed along the cylinder axis.

3. We now proceed to calculate the excited-atom effective lifetime due to the dragging of the resonant radiation. Assuming in (18) that $a_0 \sim e^{-t/\tau}$ and substituting the expression (21) for a_N , we obtain for a_0 the equation

$$\left[\gamma(1 - \hat{K}_{00}) - \gamma \sum_{N \neq 0} \frac{1}{\lambda_N} \hat{K}_{0N} \hat{K}_{N0} \right] a_0 = \frac{1}{\tau} a_0. \quad (31)$$

In the zeroth approximation, neglecting the second term in the square brackets, we obtain the usual equation for $a_0(\mathbf{r})$, an approximate solution of which was obtained by Holstein^[5]. The corresponding value τ_H of the decay time at $k_0 R \gg 1$ is

$$\tau_H = A k_0 R \sqrt{\pi \ln k_0 R} / \gamma, \quad (32)$$

where A is a numerical coefficient that depends on the shape of the vessel. The correction resulting from all types of alignment is obtained in first-order perturbation theory:

$$\Delta(\tau^{-1}) = -\gamma \sum_{N \neq 0} \frac{1}{\lambda_N} (a_0, \hat{K}_{0N} \hat{K}_{N0} a_0), \quad (33)$$

where the parentheses denote integration over the volume of the vessel, and a_0 should be taken to mean Holstein's solution so normalized that $(a_0, a_0) = 1$.

For an approximate calculation of the sum (33), we replace λ_N^{-1} by the mean value $\bar{\lambda}^{-1}$ and use the completeness of the system of the functions (3), and then the calculations lead to the following result:

$$\Delta(\tau^{-1}) = \frac{-\gamma \bar{\lambda}^{-1}}{16\pi^3} \int d^3\mathbf{r} \int a_0(\mathbf{r}_1) d^3\mathbf{r}_1 \int a_0(\mathbf{r}_2) d^3\mathbf{r}_2 \int_{-\infty}^{\infty} du_1 \int_{-\infty}^{\infty} du_2 \mathcal{F}. \quad (34)$$

Here

$$\mathcal{F} = \frac{\mathcal{F}(u_1) \mathcal{F}(u_2)}{\rho_1^2 \rho_2^2} \exp[-\mathcal{F}(u_1) \rho_1 - \mathcal{F}(u_2) \rho_2 - u_1^2 - u_2^2]$$

$$\times \left\{ \frac{B(\theta)}{\sin \theta} \exp \left[- \frac{(u_1^2 + u_2^2) \cos^2 \theta - 2u_1 u_2 \cos \theta}{\sin^2 \theta} \right] - 1 \right\}, \quad (35)$$

where $\rho_i = \mathbf{r} - \mathbf{r}_i$, θ is the angle between the vectors ρ_1 and ρ_2 , and

$$B(\theta) = \frac{3}{4} \left\{ 3 \sum_x (2j_i + 1)(2\kappa + 1) \begin{Bmatrix} \kappa & 1 & 1 \\ j_0 & j_i & j_i \end{Bmatrix}^2 \begin{Bmatrix} \kappa & 1 & 1 \\ 0 & 0 & 0 \end{Bmatrix}^2 P_{\kappa}(\cos \theta) + 1 \right\}. \quad (36)$$

The asymptotic expression of the integral (34) in the case of strong dragging ($\alpha = k_0 R \gg 1$) can be obtained by using the same procedure as in the derivation of formula (26). We then obtain for a flat layer at $j_1 = 1$ and $j_0 = 0$:

$$\Delta\tau / \tau_H = 0.9 \overline{\lambda^{-1}} / \ln \alpha, \quad (37)$$

with $\overline{\lambda^{-1}}$ a number on the order of unity.

In a magnetic field strong enough to make the distance between the Zeeman sublevels of the excited state larger than the natural level width, the alignment is destroyed. Generally speaking, however, it will not be destroyed completely, since alignment may be pre-

served along an axis coinciding with the direction of the magnetic field. Formula (37), nevertheless, gives the correct order of magnitude for the change of the effective lifetime in a magnetic field.

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