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Holes in Spectral Lines

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The decay of an atom in the presence of a static perturbation is investigated. The perturbation couples a decaying state with a nondecaying state. A "hole" appears in the emission line at a frequency equal to the frequency difference between the nondecaying state and the ground state. The effect is explained by analyzing the phase relationships of the radiation emitted by the two coupled states.

We have investigated the spontaneous emission of radiation from an atom in the presence of a static perturbation. The atom is considered to have one ground state and two excited states, one of which is nondecaying. The static perturbation couples the two excited states. Initially the atom is in the decaying state. The energy separation between the excited states is Δ and the matrix element of the static perturbation is V. The intensity of the emitted radiation is plotted in Fig. 1 as a function of the frequency for different values of V. The frequency is measured in units of γ, the decay rate of the unperturbed decaying state, with the origin of the K axis at the frequency difference between the decaying state and the ground state. The energy separation of the two excited states Δ is $0.5\,\hbar\gamma$.

For $V=0$ the emission line is Lorentzian, but for $V\neq 0$ a "hole" appears at the frequency equal to the frequency difference between the excited nondecaying state and the ground state. The position of the "hole" is independent of the static perturbation, and thus can be used to identify the nondecaying state. This effect is similar to the one discovered by Lamb[1] and subsequently used by McKibben, Lawrence, and Ohlsen[2] as a nuclear-spin filter. In Lamb's case the states are coupled by a rf field and a motional electric field. Lamb has calculated the decay constants of coupled $2\,^2P_{1/2\,1/2}$, $2\,^2S_{1/2\,-1/2}$, and $2\,^2S_{1/2\,1/2}$ states in hydrogen. At a rf equal to the frequency difference between the $2\,^2S_{1/2\,1/2}$ and $2\,^2S_{1/2\,-1/2}$ states, the rf-induced decay rate is zero. In our calculation this frequency corresponds to the "hole" frequency.

An analysis of the resonance scattering of radiation in the presence of a static perturbation also shows "holes" in the emission spectrum when a decaying state is coupled to a nondecaying state.[3] In order to be able to observe the "hole" in the frequency spectrum of the emitted radiation, the effect

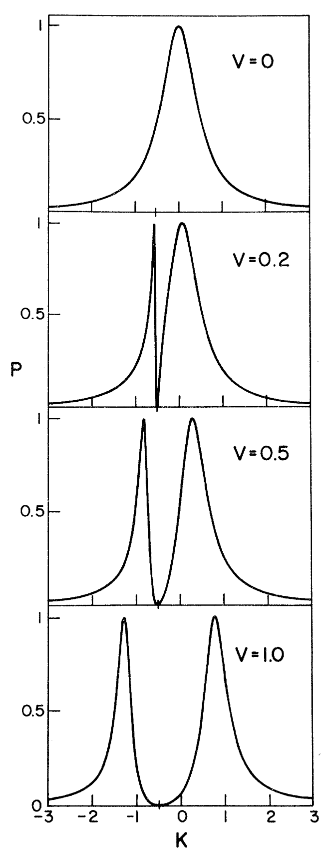

FIG. 1. Frequency distributions of emitted radiation from two coupled excited states. The frequency K is in units of γ, the natural linewidth of the unperturbed decaying state. The origin of the K axis is at the energy difference between the decaying state and the ground state. The static perturbation V is in units of $\hbar\gamma$.

of Doppler broadening has to be reduced to a minimum. This can be done by using saturation spectroscopy techniques or by detecting the light in a beam-foil experiment at right angles to the beam direction.

The appearance of the "hole" in the emission line can be more easily analyzed by assuming that the two excited states $|a\rangle$ and $|b\rangle$ are degenerate ($\Delta = 0$). The state $|a\rangle$ is coupled to a ground state $|c\rangle$ by the radiation field and state $|b\rangle$ does not decay. In the presence of the static perturbation only, the perturbed eigenstates of states $|a\rangle$ and $|b\rangle$ are

$$\begin{aligned} |1\rangle &= (1/\sqrt{2})(|a\rangle + |b\rangle), \\ |2\rangle &= (1/\sqrt{2})(|a\rangle - |b\rangle). \end{aligned} \tag{1}$$

The radiation field now couples both states $|1\rangle$ and $|2\rangle$ to the ground state $|c\rangle$. In the interaction representation the state vector of the system at any time t is given by

$$|\psi(t)\rangle = b_1(t)|1\rangle + b_2(t)|2\rangle + \sum_f b_f(t)|f\rangle, \tag{2}$$

where

$$|f\rangle = |c\rangle|k_\lambda\rangle .$$

The summation over f is over frequency, direction, and polarization of the emitted radiation. The state $|k_\lambda\rangle$ represents a photon with wave vector $\vec{k}_\lambda$ and polarization λ. The time-dependent coefficients in Eq. (2) satisfy the following differential equations[4]:

$$\begin{aligned} i\hbar\dot{b}_1(t) &= \sum_f H_{1f} b_f(t) e^{i(E_1-E_f)t/\hbar} + i\hbar A_1\delta(t), \\ i\hbar\dot{b}_2(t) &= \sum_f H_{2f} b_f(t) e^{i(E_2-E_f)t/\hbar} + i\hbar A_2\delta(t), \\ i\hbar\dot{b}_f(t) &= H_{f1} b_1(t) e^{i(E_f-E_1)t/\hbar} + H_{f2} b_2(t) e^{i(E_f-E_2)t/\hbar}. \end{aligned} \tag{3}$$

The coefficients A_1 and A_2 are the initial amplitudes of the states $|1\rangle$ and $|2\rangle$, respectively. For the atom to be in state $|a\rangle$ at $t=0$ requires $A_1 = A_2 = 1/\sqrt{2}$. In Eq. (3), E_1 and E_2 are the energies of the eigenstates $|1\rangle$ and $|2\rangle$, respectively, and $E_f = E_c + \hbar c k_\lambda$. The matrix elements of the interaction of the atom with the radiation field are

$$\begin{aligned} H_{jf} &= \frac{e}{mc}\langle j|\vec{p}\cdot\vec{A}|f\rangle \\ &= \frac{e}{mc}\frac{1}{\sqrt{2}}\langle a|\vec{p}\cdot\vec{A}|f\rangle, \quad j=1,2 . \end{aligned}$$

The differential equations can be solved by Fourier-transforming the expansion coefficients[4,5]:

$$b_n(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} dE\, G_n e^{i(E_n-E)t/\hbar} . \tag{4}$$

This leads to a set of linear equations for the Fourier coefficients:

$$\begin{aligned} (E-E_1)G_1 &= \sum_f H_{1f}G_f + 1/\sqrt{2}, \\ (E-E_2)G_2 &= \sum_f H_{2f}G_f + 1/\sqrt{2}, \\ (E-E_f)G_f &= H_{f1}G_1 + H_{f2}G_2 . \end{aligned} \tag{5}$$

These equations are solved by first eliminating the Fourier coefficients of the final states G_f and by defining

$$\sum_f H_{jf}H_{fl}\,\zeta(E-E_f) = -\tfrac{1}{4}i\hbar\gamma \quad (j,\ l=1,2),$$

where

$$-\tfrac{1}{2}i\hbar\gamma = (e/mc)^2\sum_f \langle a|\vec{p}\cdot\vec{A}|f\rangle\langle f|\vec{p}\cdot\vec{A}|a\rangle\,\zeta(E-E_f).$$

The following representation of the zeta function is used:

$$\zeta(E-E_f) = \mathcal{P}\left(\frac{1}{E-E_f}\right) - i\pi\delta(E-E_f),$$

where $\mathcal{P}$ is the principal-value operator. The real part of γ is the decay constant of state $|a\rangle$. It is practically independent of E.[6] The imaginary part of γ produces an energy shift of the atomic levels. If a representation is used where E_1 and E_2 include the self-energies, then the imaginary part of γ vanishes. In the following calculation it is assumed that the energies are renormalized.

The solutions of Eq. (5) are

$$\begin{aligned} G_1 &= \frac{E-E_2}{\sqrt{2}\,((E-E_1+\frac{1}{4}i\hbar\gamma)(E-E_2+\frac{1}{4}i\hbar\gamma)+\frac{1}{16}\hbar^2\gamma^2)}, \\ G_2 &= \frac{E-E_1}{\sqrt{2}\,((E-E_1+\frac{1}{4}i\hbar\gamma)(E-E_2+\frac{1}{4}i\hbar\gamma)+\frac{1}{16}\hbar^2\gamma^2)}, \\ G_f &= \frac{[H_{f1}(E-E_2)+H_{f2}(E-E_1)]\zeta(E-E_f)}{\sqrt{2}\,((E-E_1+\frac{1}{4}i\hbar\gamma)(E-E_2+\frac{1}{4}i\hbar\gamma)+\frac{1}{16}\hbar^2\gamma^2)} . \end{aligned} \tag{6}$$

The real parts of the poles of G_f are the energies of the perturbed system and the imaginary parts are the decay constants. The poles are at

$$\begin{aligned} \left.\begin{matrix} E_1' \\ E_2' \end{matrix}\right\} &= \tfrac{1}{2}(E_1+E_2) \pm \tfrac{1}{2}(4V^2 - \tfrac{1}{4}\hbar^2\gamma^2)^{1/2} - \tfrac{1}{4}i\hbar\gamma, \\ E_3' &= E_f, \end{aligned} \tag{7}$$

where $V^2 = |\langle a|V|b\rangle|^2$.

For $V^2 > \frac{1}{16}\hbar^2\gamma^2$, the square root in Eq. (7) contributes to the energies of the perturbed states, whereas for $V^2 < \frac{1}{16}\hbar^2\gamma^2$ the square root affects the decay of the system.

The amplitude of the final states is now obtained by contour integration:

$$b_f(t) = (\sqrt{2})H_{f1}\left(\frac{E_f - \frac{1}{2}(E_1+E_2)}{(E_f-E_1')(E_f-E_2')} + \frac{E_1' - \frac{1}{2}(E_1+E_2)}{(E_1'-E_f)(E_1'-E_2')}\,e^{i(E_f-E_1')t/\hbar}\right.$$

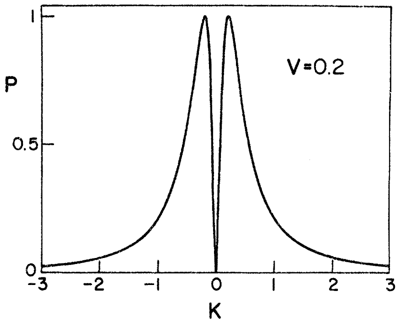

FIG. 2. Frequency distribution of emitted radiation from two coupled excited states. The unperturbed states are degenerate. The frequency K is in units of γ, the natural linewidth of the unperturbed decaying state. The origin of the K axis is at the energy difference between the unperturbed excited states and the ground state. The static perturbation is in units of $\hbar\gamma$.

$$+\frac{E_2'-\frac{1}{2}(E_1+E_2)}{(E_2'-E_f)(E_2'-E_1')}\, e^{i(E_f-E_2')t/\hbar}\Bigg), \tag{8}$$

and as $t\to\infty$ it reduces to

$$b_f(\infty)=(\surd 2)\,H_{f1}\,\frac{E_f-\frac{1}{2}(E_1+E_2)}{(E_f-E_1')(E_f-E_2')}\ . \tag{9}$$

Thus the probability of emission of a photon of wave vector $\vec{k}_\lambda$ and polarization λ is

$$|b_f(\infty)|^2=\frac{2|H_{f1}|^2}{\hbar^2\gamma^2}\left|\frac{K}{(K-R+\frac{1}{4}i)(K+R+\frac{1}{4}i)}\right|^2, \tag{10}$$

where

$$K=[\hbar c k_\lambda-\tfrac{1}{2}(E_1+E_2)+E_c]/\hbar\gamma,$$

$$R=(1/2\hbar\gamma)(4V^2-\tfrac{1}{4}\hbar^2\gamma^2)^{1/2}\ .$$

This probability is plotted in Fig. 2 as a function of the frequency of the emitted photon. A "hole" appears at a frequency equal to the frequency difference of the nondecaying excited state and the ground state.

The appearance of the "hole" can be investigated in more detail by considering the time dependence of the amplitude at the "hole" frequency ($K=0$). This amplitude is given by

$$b_f(t)=\frac{H_{f1}}{(\surd 2)\hbar\gamma R}\,(e^{-iR\gamma t}-e^{iR\gamma t})\,e^{-\gamma t/4}\ . \tag{11}$$

For $|V|>\frac{1}{4}\hbar\gamma$ the probability can be written as

$$|b_f(t)|^2=\frac{2|H_{f1}|^2}{\hbar^2\gamma^2R^2}\sin^2(R\gamma t)e^{-\gamma t/2}\ , \tag{12}$$

which is zero for $t=n\pi/\gamma R$ ($n=0,\ 1,\ 2,\ \ldots$) and for $t\to\infty$. For $|V|<\frac{1}{4}\hbar\gamma$ the probability of photon emission at $K=0$ is

$$|b_f(t)|^2=\frac{2|H_{f1}|^2}{\hbar^2\gamma^2|R|^2}\sinh^2(|R|\gamma t)\,e^{-\gamma t/2}\ , \tag{13}$$

which vanishes only for $t=0$ and for $t\to\infty$.

This probability is plotted in Fig. 3 for $V=0.2\hbar\gamma$ and for $V=\hbar\gamma$. It is the probability that a photon has been emitted at time t. This interpretation, however, causes difficulties when trying to explain the decreasing probability at certain times. It is particularly apparent for $V=0.2\hbar\gamma$, where the probability continually decreases for t greater than $4.62\gamma^{-1}$. This would lead to negative currents. If, however, it is assumed that the atom emits radiation continuously, then the probability becomes the detector response at the "hole" frequency of a pulse of duration t. The longer the pulse, the less the detector responds at this frequency.

One can obtain the amplitude of emitted radiation at $K=0$ by separating the contributions from states $|1\rangle$ and $|2\rangle$. They are, respectively,

$$b_{f1}(t)=\frac{H_{f1}}{(\surd 2)\hbar\gamma}\Bigg(-\frac{V}{R^2+\frac{1}{16}}+\frac{V+R-\frac{1}{4}i}{2R(R-\frac{1}{4}i)}\times e^{-iR\gamma t-\gamma t/4}+\frac{V-R-\frac{1}{4}i}{2R(R+\frac{1}{4}i)}\,e^{iR\gamma t-\gamma t/4}\Bigg) \tag{14}$$

and

$$b_{f2}(t)=\frac{H_{f2}}{(\surd 2)\hbar\gamma}\Bigg(\frac{V}{R^2+\frac{1}{16}}+\frac{R-V-\frac{1}{4}i}{2R(R-\frac{1}{4}i)}\,e^{-iR\gamma t-\gamma t/4}-\frac{R+V+\frac{1}{4}i}{2R(R+\frac{1}{4}i)}\,e^{iR\gamma t-\gamma t/4}\Bigg)\ . \tag{15}$$

For $|V|>\frac{1}{4}\hbar\gamma$ the two contributions reduce to

$$b_{f1}(t)=\frac{H_{f1}}{(\surd 2)\hbar\gamma R(R^2+\frac{1}{16})}\,[-RV+RV\cos(R\gamma t)\,e^{-\gamma t/4}-i(R^2+\tfrac{1}{16}+\tfrac{1}{4}iV)\sin(R\gamma t)\,e^{-\gamma t/4}] \tag{16}$$

and

$$b_{f2}(t)=\frac{H_{f1}}{(\surd 2)\hbar\gamma R(R^2+\frac{1}{16})}\,[RV-RV\cos(R\gamma t)e^{-\gamma t/4}$$

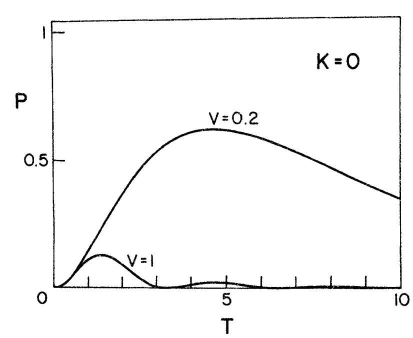

FIG. 3. Probabilities of photon emission as a function of time. The frequency corresponds to the energy difference between the unperturbed degenerate excited states and the ground state. The time is measured in units of γ^{-1} and the static perturbation in units of $\hbar\gamma$, where γ^{-1} is the lifetime of the unperturbed decaying state.

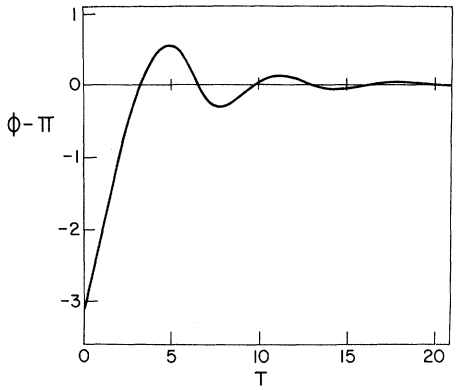

FIG. 4. Phase difference ϕ between the oscillators 1 and 2 as a function of time at $K=0$ and $V=1$. The time is measured in units of γ^{-1}, and the static perturbation V in units of $\hbar\gamma$.

$$-i(R^2+\tfrac{1}{16}-\tfrac{1}{4}iV)\sin(R\gamma t)e^{-\gamma t/4}\,] \,. \quad (17)$$

If one writes these amplitudes as

$$b_{f1}(t)=\alpha e^{i\theta_1}\,, \quad b_{f2}(t)=\beta e^{i\theta_2}\,, \quad (18)$$

one finds that $\alpha=\beta$ and

$$\cos\theta_1=-\cos\theta_2\,, \quad \sin\theta_1=\sin\theta_2\,. \quad (19)$$

The phase relationships can be written as

$$\theta_1(t)+\theta_2(t)=(2n+1)\pi \quad (n=0,\ 1,\ 2,\ \ldots)\,. \quad (20)$$

The condition that the contributions to $b_f(t)$ are out of phase gives

$$\theta_1(t)-\theta_2(t)=(2m+1)\pi \quad (m=0,\ 1,\ 2,\ \ldots)\,,$$

which together with the general phase relationship yields

$$\theta_1(t)=(n+m+1)\pi, \quad \theta_2(t)=(n-m)\pi\,.$$

This implies that if for a given time θ_1 is an odd multiple of π, then θ_2 is an even multiple of π and the two contributions are out of phase and thus the total amplitude is zero. The two radiators 1 and 2 interfere destructively at $K=0$, at times $t=l\pi/\gamma R$ ($l=1, 2, \ldots$), and as $t\to\infty$.

The phase difference $\phi=\theta_1-\theta_2$ can be evaluated for arbitrary times in the following way.

From Eqs. (18) and (19) the sum of the amplitudes $b_{f1}(t)$ and $b_{f2}(t)$ yields

$$b_f(t)=b_{f1}(t)+b_{f2}(t)=2i\alpha\sin(\theta_1)\,, \quad (21)$$

and by comparing Eq. (21) with Eq. (11) one gets

$$\theta_1=\sin^{-1}\left(-\frac{H_{f1}}{(\sqrt{2})\hbar\gamma R\alpha}\sin(R\gamma t)e^{-\gamma t/4}\right). \quad (22)$$

The phase difference ϕ can now be written in terms of the angle θ_1 by using the relation between θ_1 and θ_2 from Eq. (20). The result is

$$\phi=2\sin^{-1}\left(-\frac{H_{f1}}{(\sqrt{2})\hbar\gamma R\alpha}\sin(R\gamma t)e^{-\gamma t/4}\right)+\pi\,.$$

Figure 4 shows this phase difference as a function of time. At $t=0$ the two oscillators are in phase; at $t=3.245\gamma^{-1}$ they are out of phase, and as time increases, the deviation from out of phase becomes less and less.

It is interesting to note that the "hole" disappears if the atom is in the nondecaying excited state at $t=0$. For $|V|>\frac{1}{4}\hbar\gamma$ and $\Delta=0$, the emission line has two peaks which are separated by $2|V|$. For $|V|<\frac{1}{4}\hbar\gamma$ and $\Delta=0$, one sharp peak at $K=0$ appears and the width of this line decreases as the external static perturbation decreases.

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