Theoretical investigation of energy-trapping mechanism by atomic systems

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The theoretical results are presented here in detail for the atomic device proposed earlier by the author. This device absorbs energy from a continuous radiation source and stores some of it with atoms in metastable states for a long time without any loss. At a later time, when the energy is required, the system can be "triggered" by an external perturbing field to release the energy in the form of a strong pulse of radiation.

I. INTRODUCTION

In an earlier Letter[1] the author has proposed an atomic device for trapping energy from continuous radiation. Only a brief introduction to the device and its working mechanism was given there. The purpose of this paper is to present a complete theoretical analysis of the device and to discuss its possible applications.

The device consists of a three-level atomic gas[2] with two excited states. One of the excited states does *not* decay to the ground state through emission of radiation (we shall consider only single-photon processes). The atomic gas is irradiated by continuous radiation in the presence of an external perturbing field[3] which couples the two excited states. The perturbation causes the atoms to be populated in both the upper states after absorbing the energy form the incident radiation. These atoms at once start emitting radiation through a resonance-fluorescence process.[4] But, before this process is complete, the perturbation field is turned off, which leaves the atom with a finite probability of being in the nondecaying (metastable) state. Thus, a finite amount of energy from the incident radiation has been *trapped* with atoms in their metastable states. This mechanism is discussed in Sec. II in detail for an exponentially decaying field.[5]

The *trapped* energy can be stored for a long time (as long as the lifetime of the metastable state) and can be released at any time in the form of radiation by *triggering* the device through a static perturbation which again couples the two excited states. This is further discussed in Sec. III. Since the total trapped energy is emitted in a very short time, the released pulse is expected to have huge power. This aspect along with some of the possible applications is discussed in Sec. IV.

II. ENERGY-TRAPPING MECHANISM

Here we want to find the probability of an atom being in the nondecaying excited state after absorbing the energy from the incident radiation in the presence of a short-lived coupling field. This is achieved by solving the Schrödinger equation of motion for the system. Initially, the atom is considered to be in the ground state $|c\rangle$ and a beam of continuous radiation is incident on it. The two excited states $|a\rangle$ and $|b\rangle$, with $|a\rangle$ the upper one, are coupled by an external time-dependent potential of the form

$$V(\vec{r},t)=V(\vec{r})e^{-\epsilon t}, \quad t\geqslant 0 \tag{1}$$

where $V(\vec{r})$ depends on the nature of the coupling field. For example, in the case of an electric field, this would represent the electric-dipole interaction.

The excited state $|b\rangle$ is assumed to be a nondecaying (metastable) state. No coupling is considered between the ground state and the excited states through the perturbation field.

The Schrödinger equation of motion for the system is

$$i\hbar\frac{\partial}{\partial t}\,|\psi(t)\rangle=[H_0+H+V(t)]\,|\psi(t)\rangle\,, \tag{2}$$

where H_0 is the unpertrubed Hamiltonian of the atom and the radiation field, and H represents the interaction of the atom with the radiation field. This interaction, in the absence of multiphoton processes, consists of only $\vec{A}\cdot\vec{p}$ terms,[6] where $\vec{A}$ represents the vector potential of the radiation field and $\vec{p}$ is the linear momentum of the electron in the atom.

The state vector $|\psi(t)\rangle$ of the system at any time t can be written as a linear combination of the eigenvectors of the unperturbed Hamiltonian H_0 of the system:

$$|\psi(t)\rangle=c_0(t)\,|c;0\rangle e^{-i\omega_0 t}+\sum_{\vec{k}\sigma}a_{\vec{k}\sigma}(t)\,|a;-\vec{k}\sigma\rangle\,e^{-i\omega_{a\sigma}t}+\sum_{\vec{k}\sigma}b_{\vec{k}\sigma}(t)\,|b;-\vec{k}\sigma\rangle e^{-i\omega_{b\sigma}t}$$
$$+\sum_{\vec{k}\sigma\vec{k}'\lambda}c_{\vec{k}\sigma\vec{k}'\lambda}(t)\,|c;-\vec{k}\sigma;\vec{k}'\lambda\rangle\,e^{-i\omega_{c\sigma\lambda}t}\quad . \tag{3}$$

The symbols $c_0(t)$, $a_{\vec{k}\sigma}(t)$, $b_{\vec{k}\sigma}(t)$, and $c_{\vec{k}\sigma\vec{k}'\lambda}(t)$ represent the probability amplitudes in the corresponding states which are given below in terms of the product functions of the unperturbed atomic and photon states:

$$\begin{aligned} |c;0\rangle &\equiv |c\rangle\,|0\rangle_{\rm rad}\,,\\ |a;-\vec{k}\sigma\rangle &\equiv |a\rangle\,|-\vec{k}\sigma\rangle_{\rm rad}\,,\\ |b;-\vec{k}\sigma\rangle &\equiv |b\rangle\,|-\vec{k}\sigma\rangle_{\rm rad}\,,\\ |c;-\vec{k}\sigma;\vec{k}'\lambda\rangle &\equiv |c\rangle|-\vec{k}\sigma\rangle_{\rm rad}\,|\vec{k}'\lambda\rangle_{\rm rad}\,. \end{aligned} \tag{4}$$

The state $|0\rangle_{\rm rad}$ represents the initial state of the radiation field with no photons being absorbed whereas $|-\vec{k}\sigma\rangle_{\rm rad}$ and $|\vec{k}'\lambda\rangle_{\rm rad}$ represent the states of the field with an absorbed photon of wave vector $\vec{k}$, polarization σ, and frequency ω_σ, and an emitted photon of wave vector $\vec{k}'$, polarization λ, and frequency ω_λ, respectively. In general, these states are written in terms of the Fock states as $|\cdots N_{\vec{k}\lambda}, N_{\vec{k}'\lambda'}, N_{\vec{k}''\lambda''}\cdots\rangle$, where the $N_{\vec{k}\lambda}$'s represent the numbers of photons with their corresponding wave vectors $\vec{k}$ and polarizations λ. However, this explicit form becomes very cumbersome to write; therefore, the states in Eq. (4) are normally preferred. Moreover, in the radiation problems, we mostly deal with the energy changes during the absorption or emission processes; therefore, one can assume that the initial energy of the radiation field is zero. This is the case for the photon states defined in Eq. (4), though the detailed forms of the states are required in computing the matrix elements of the interaction Hamiltonian H.

The eigenfrequencies in Eq. (3) are defined as

$$\begin{aligned} &\omega_0 = E_c/\hbar, \quad \omega_{a\sigma} = E_a/\hbar - \omega_\sigma\,,\\ &\omega_{b\sigma} = E_b/\hbar - \omega_\sigma, \quad \omega_{c\sigma\lambda} = E_c/\hbar - \omega_\sigma + \omega_\lambda\,, \end{aligned} \tag{5}$$

where E_a, E_b, and E_c are the energies of the atom in states $|a\rangle$, $|b\rangle$, and $|c\rangle$, respectively.

Combining Eqs. (1)–(3) we obtain the following equations of motion for the probability amplitudes:

$$i\hbar\dot{c}_0(t) = \sum_{\vec{k}\sigma} H_{0;a\vec{k}\sigma}\, a_{\vec{k}\sigma}(t)\exp[i(\omega_0 - \omega_{a\sigma})t]\,, \tag{6}$$

$$\begin{aligned} i\hbar\dot{a}_{\vec{k}\sigma}(t) = {} & H_{a\vec{k}\sigma;0}\, c_0(t)\exp[i(\omega_{a\sigma} - \omega_0)t]\\ & + \sum_{\vec{k}'\lambda} H_{a\vec{k}\sigma;c\vec{k}\sigma\vec{k}'\lambda}\, c_{\vec{k}\sigma\vec{k}'\lambda}(t)\exp[i(\omega_{a\sigma} - \omega_{c\sigma\lambda})t]\\ & + V_{a\vec{k}\sigma;b\vec{k}\sigma}\, b_{\vec{k}\sigma}(t)\exp[-\epsilon t + i(\omega_{a\sigma} - \omega_{b\sigma})t]\,, \end{aligned} \tag{7}$$

$$\begin{aligned} i\hbar\dot{b}_{\vec{k}\sigma}(t) = {} & V_{b\vec{k}\sigma;a\vec{k}\sigma}\, a_{\vec{k}\sigma}(t)\\ & \times\exp[-\epsilon t + i(\omega_{b\sigma} - \omega_{a\sigma})t]\,, \end{aligned} \tag{8}$$

$$\begin{aligned} i\hbar\dot{c}_{\vec{k}\sigma\vec{k}'\lambda}(t) = {} & H_{c\vec{k}\sigma\vec{k}'\lambda;a\vec{k}\sigma}\, a_{\vec{k}\sigma}(t)\\ & \times\exp[i(\omega_{c\sigma\lambda} - \omega_{a\sigma})t]\,, \end{aligned} \tag{9}$$

where the different matrix elements in the above equations are defined as

$$H_{a\vec{k}\sigma;0} \equiv \langle a;\vec{k}\sigma\,|H|\,c;0\rangle\,, \tag{10}$$

$$H_{c\vec{k}\sigma\vec{k}'\lambda;a\vec{k}\sigma} \equiv \langle c;-\vec{k}\sigma;\vec{k}'\lambda\,|H|\,a;-\vec{k}\sigma\rangle\,, \tag{11}$$

and

$$\begin{aligned} V_{a\vec{k}\sigma;b\vec{k}\sigma} &\equiv \langle a;-\vec{k}\sigma\,|V(\vec{r})|\,b;-\vec{k}\sigma\rangle\\ &= \langle a\,|V(\vec{r})|\,b\rangle\,. \end{aligned} \tag{12}$$

Here, it is assumed that the states $|a\rangle$ and $|b\rangle$ are not coupled by the radiation interaction H and neither are the states $|b\rangle$ and $|c\rangle$ as considered earlier. The diagonal matrix elements of $V(\vec{r})$ are assumed to be zero. Although, if they were not zero, one could absorb them in the respective energies of the atomic states, and the final equations [Eqs. (21)–(24)] to be solved would not look any different.

By making the following substitutions for the probability amplitudes

$$A_{\vec{k}\sigma}(t) \equiv a_{\vec{k}\sigma}(t)\exp[i(\omega_0 - \omega_{a\sigma})t]\,, \tag{13}$$

$$B_{\vec{k}\sigma}(t) \equiv b_{\vec{k}\sigma}(t)\exp[i(\omega_0 - \omega_{b\sigma})t]\,, \tag{14}$$

$$C_{\vec{k}\sigma\vec{k}'\lambda}(t) \equiv c_{\vec{k}\sigma\vec{k}'\lambda}(t)\exp[i(\omega_0 - \omega_{c\sigma\lambda})t]\,, \tag{15}$$

and

$$C_0(t) \equiv c_0(t) \tag{16}$$

in Eqs. (6)–(9), one obtains

$$i\hbar\dot{C}_0(t) = \sum_{\vec{k}\sigma} H_{0;a\vec{k}\sigma}\, A_{\vec{k}\sigma}(t)\,, \tag{17}$$

$$\begin{aligned} i\hbar[\dot{A}_{\vec{k}\sigma}(t) + i(\omega_{a\sigma} - \omega_0)A_{\vec{k}\sigma}(t)] &= H_{a\vec{k}\sigma;0}C_0(t) + \sum_{\vec{k}'\lambda} H_{a\vec{k}\sigma;c\vec{k}\sigma\vec{k}'\lambda}C_{\vec{k}\sigma\vec{k}'\lambda}(t)\\ &\quad + V_{ab}B_{\vec{k}\sigma}(t)e^{-\epsilon t}\,, \end{aligned} \tag{18}$$

$$i\hbar[\dot{B}_{\vec{k}\sigma}(t) + i(\omega_{b\sigma} - \omega_0)B_{\vec{k}\sigma}(t)] = V_{ba}A_{\vec{k}\sigma}(t)e^{-\epsilon t}\,, \tag{19}$$

$$\begin{aligned} i\hbar[\dot{C}_{\vec{k}\sigma\vec{k}'\lambda}(t) + i(\omega_{c\sigma\lambda} - \omega_0)C_{\vec{k}\sigma\vec{k}'\lambda}(t)]\\ = H_{c\vec{k}\sigma\vec{k}'\lambda;a\vec{k}\sigma}A_{\vec{k}\sigma}(t)\,, \end{aligned} \tag{20}$$

where $V_{ab} = \langle a\,|\,V(\vec{r})\,|\,b\rangle$.

In order to solve for the probability amplitudes, we Laplace transform Eqs. (17)–(20). Using the initial condition that $C_0(0) = 1$ and the other amplitudes are zero at $t = 0$, we get

$$sY_0(s) = 1 - i\sum_{\vec{k}\sigma}(H_{0;a\vec{k}\sigma}/\hbar)Y_{a\vec{k}\sigma}(s)\,, \tag{21}$$

$$\begin{aligned} [s + i(\omega_{a\sigma} - \omega_0)]Y_{a\vec{k}\sigma}(s) = {} & -i\frac{H_{a\vec{k}\sigma;0}}{\hbar}Y_0(s)\\ & - i\sum_{\vec{k}'\lambda}\frac{H_{a\vec{k}\sigma;c\vec{k}\sigma\vec{k}'\lambda}}{\hbar}Y_{c\vec{k}\sigma\vec{k}'\lambda}(s)\\ & - i(V_{ab}/\hbar)Y_{b\vec{k}\sigma}(s+\epsilon)\,, \end{aligned} \tag{22}$$

$$[s+i(\omega_{b\sigma}-\omega_0)]Y_{b\vec{k}\sigma}(s)=-i\,\frac{V_{ba}}{\hbar}\,Y_{a\vec{k}\sigma}(s+\epsilon)\,, \tag{23}$$

$$[s+i(\omega_{c\sigma\lambda}-\omega_0)]Y_{c\vec{k}\sigma\vec{k}'\lambda}(s)=-i(H_{c\vec{k}\sigma\vec{k}'\lambda;a\vec{k}\sigma}/\hbar)Y_{a\vec{k}\sigma}(s)\,, \tag{24}$$

where the $Y(s)$'s represent the Laplace transforms of the new amplitudes defined in Eqs. (13)–(16):

$$Y_n(s)=\int_0^\infty e^{-st}A_n(t)\,dt\,. \tag{25}$$

Combining Eqs. (22)–(24) yields

$$Y_{a\vec{k}\sigma}(s)=[s+\tfrac{1}{2}\gamma(s)+i(\omega_{a\sigma}-\omega_0)]^{-1}\times\{-i(H_{a\vec{k}\sigma;0}/\hbar)Y_0(s)-|V_{ab}/\hbar|^2\times Y_{a\vec{k}\sigma}(s+2\epsilon)[s+\epsilon+i(\omega_{b\sigma}-\omega_0)]^{-1}\}\,, \tag{26}$$

where

$$\tfrac{1}{2}\gamma(s)=\sum_{\vec{k}'\lambda}\frac{|H_{a\vec{k}\sigma;c\vec{k}\sigma\vec{k}'\lambda}/\hbar|^2}{s+i(\omega_\lambda-\omega_\sigma)}\,. \tag{27}$$

Although γ is a function of s, it can be shown[7,8] that its real part is independent of s and represents the decay constant of the unperturbed state $|a\rangle$. The imaginary part of γ, which diverges, represents the shift in the energy of the state. When the energies are renormalized by considering the self-energies of the state, the shift vanishes and γ becomes real. Thus, hereafter, γ is considered to be real and a constant representing the decay constant of the state $|a\rangle$ in the absence of perturbation.

The exact solution of $Y_{a\vec{k}\sigma}(s)$ from Eq. (26) is not possible. Therefore, an approximate method suggested by Fontana and Thomann[9] has been used. When the argument s in Eq. (26) is replaced by $s+2\epsilon$, one obtains an expression for $Y_{a\vec{k}\sigma}(s+2\epsilon)$ in terms of $Y_0(s+2\epsilon)$ and $Y_{a\vec{k}\sigma}(s+4\epsilon)$. Similarly, one can write $Y_{a\vec{k}\sigma}(s+4\epsilon)$ in terms of $Y_0(s+4\epsilon)$ and $Y_{a\vec{k}\sigma}(s+6\epsilon)$. Using this iterative procedure, we can write the second-order solution, by neglecting $Y_{a\vec{k}\sigma}(s+6\epsilon)$, as

$$\begin{aligned}Y^{(2)}_{a\vec{k}\sigma}(s)=&-i(H_{a\vec{k}\sigma;0}/\hbar)[s+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]^{-1}\\&\times(Y_0(s)-|V_{ab}/\hbar|^2Y_0(s+2\epsilon)\{[s+\epsilon+i(\omega_{b\sigma}-\omega_0)][s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1}+|V_{ab}/\hbar|^4Y_0(s+4\epsilon)\\&\times\{[s+\epsilon+i(\omega_{b\sigma}-\omega_0)][s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)][s+3\epsilon+i(\omega_{b\sigma}-\omega_0)][s+4\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1})\,.\end{aligned} \tag{28}$$

This expression is substituted in Eq. (21) which yields the following recurrence relation for $Y_0(s)$:

$$\begin{aligned}Y_0(s)=[s+\tfrac{1}{2}\Gamma(s)]^{-1}\Bigg(1+\left|\frac{V_{ab}}{\hbar}\right|^2Y_0(s+2\epsilon)&\\\times\sum_{\vec{k}\sigma}\left|\frac{H_{0;a\vec{k}\sigma}}{\hbar}\right|^2\{[s+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)][s+\epsilon+i(\omega_{b\sigma}-\omega_0)]&\\\times[s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1}-\left|\frac{V_{ab}}{\hbar}\right|^4Y_0(s+4\epsilon)&\\\times\sum_{\vec{k}\sigma}\left|\frac{H_{0;a\vec{k}\sigma}}{\hbar}\right|^2\{[s+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)][s+\epsilon+i(\omega_{b\sigma}-\omega_0)]&\\\times[s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)][s+3\epsilon+i(\omega_{b\sigma}-\omega_0)]&\\\times[s+4\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1}\Bigg)\,,&\end{aligned} \tag{29}$$

where

$$\tfrac{1}{2}\Gamma(s)=\sum_{\vec{k}\sigma}\frac{|H_{0;a\vec{k}\sigma}/\hbar|^2}{s+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)}\,. \tag{30}$$

In general, Γ appears to be a function of s as was γ in Eq. (27). However, it can be shown[7,8] again that the real part of Γ is independent of s and represents the decay constant of the initial state. The imaginary part of Γ diverges and can be neglected in the representation of renormalized states as discussed earlier. Therefore, hereafter, $\Gamma(s)$ is assumed to be real and equal to a constant Γ.[10]

The summations over $\vec{k}$ in Eq. (29) are replaced by integrals over the frequency ω_σ of the incident photon ($\sum_{\vec{k}}\to\int\rho(\omega_\sigma)\,d\omega_\sigma$; $\rho(\omega_\sigma)$ being the density of states. [See Eq. (16) on p. 179 and Eq. (14) on p. 200 of Ref. 7]. The resulting integrals are evaluated by contour integration. The main contribution comes from the poles. Since $\rho(\omega_\sigma)\,|H_{0;a\vec{k}\sigma}(\omega_\sigma)|^2$ in the integrals is a slowly varying function of ω_σ near the poles, one can easily replace it by a constant. The limits of integration for ω_σ can, now, be extended from $0\to\infty$

to $-\infty \to \infty$ without significant error. The integrals, when evaluation through contour integration, yield zero contribution. Therefore, Eq. (29) reduces to

$$Y_0(s) = (s + \tfrac{1}{2}\Gamma)^{-1} . \tag{31}$$

Here, Γ represents the decay constant of the initial state as mentioned earlier. It is interesting to see that the decay of the initial state does not depend on the external field which couples the two excited states.

The second-order solution of $Y_{a\vec{k}\sigma}(s)$ can now be obtained by substituting $Y_0(s)$ from Eq. (31) into Eq. (28):

$$\begin{aligned} Y^{(2)}_{a\vec{k}\sigma}(s) = &-i(H_{a\vec{k}\sigma;0}/\hbar)[s+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]^{-1} \\ &\times\big([s+\tfrac{1}{2}\Gamma]^{-1} - |V_{ab}/\hbar|^2\{[s+2\epsilon+\tfrac{1}{2}\Gamma][s+\epsilon+i(\omega_{b\sigma}-\omega_0)][s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1} \\ &+|V_{ab}/\hbar|^4\{[s+4\epsilon+\tfrac{1}{2}\Gamma][s+\epsilon+i(\omega_{b\sigma}-\omega_0)][s+2\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)][s+3\epsilon+i(\omega_{b\sigma}-\omega_0)] \\ &\times[s+4\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1}\big) . \end{aligned} \tag{32}$$

Similarly, for $Y_{b\vec{k}\sigma}(s)$, we obtain from Eqs. (23) and (32)

$$\begin{aligned} Y^{(2)}_{b\vec{k}\sigma}(s) = &-(V_{ba}H_{a\vec{k}\sigma;0}/\hbar^2)\{[s+i(\omega_{b\sigma}-\omega_0)][s+\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1} \\ &\times\big((s+\epsilon+\tfrac{1}{2}\Gamma)^{-1} - |V_{ab}/\hbar|^2\{(s+3\epsilon+\tfrac{1}{2}\Gamma)[s+2\epsilon+i(\omega_{b\sigma}-\omega_0)][s+3\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1} \\ &+|V_{ab}/\hbar|^4\{(s+5\epsilon+\tfrac{1}{2}\Gamma)[s+2\epsilon+i(\omega_{b\sigma}-\omega_0)][s+3\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)] \\ &\times[s+4\epsilon+i(\omega_{b\sigma}-\omega_0)][s+5\epsilon+\tfrac{1}{2}\gamma+i(\omega_{a\sigma}-\omega_0)]\}^{-1}\big) . \end{aligned} \tag{33}$$

The time-dependent amplitudes can now be obtained by the inverse Laplace transforms of the $Y(s)$'s. In fact, the $Y(s)$'s are the Laplace transforms of the redefined amplitudes in Eqs. (13)–(16). However, they are directly related to the Laplace transforms $y(s)$ of the original amplitudes (defined in Eq. 3) by

$$y_n(s) = Y_n[s - i(\omega_n - \omega_0)] . \tag{34}$$

In the present paper, we are mainly interested in the probability amplitudes at $t\to\infty$. These can be easily determined by using the final-value theorem[11] which yields

$$\begin{aligned} b^{(2)}_{\vec{k}\sigma}(\infty) &= \lim_{s\to 0}[sy^{(2)}_{b\vec{k}}(s)] = \lim_{s\to 0}\{sY^{(2)}_{b\vec{k}\sigma}[s-i(\omega_{b\sigma}-\omega_0)]\} \\ &= -(V_{ba}H_{a\vec{k}\sigma;0}/\hbar^2)[\omega_{ba}+i(\epsilon+\tfrac{1}{2}\gamma)]^{-1} \\ &\quad\times\big([\omega_\sigma-\omega_{bc}-i(\epsilon+\tfrac{1}{2}\Gamma)]^{-1} - i\,|V_{ab}/\hbar|^2\{2\epsilon[\omega_{ba}+i(3\epsilon+\tfrac{1}{2}\gamma)][\omega_\sigma-\omega_{bc}-i(3\epsilon+\tfrac{1}{2}\Gamma)]\}^{-1} \\ &\quad-|V_{ab}/\hbar|^4\{8\epsilon^2[\omega_{ba}+i(3\epsilon+\tfrac{1}{2}\gamma)][\omega_{ba}+i(5\epsilon+\tfrac{1}{2}\gamma)][\omega_\sigma-\omega_{bc}-i(5\epsilon+\tfrac{1}{2}\Gamma)]\}^{-1}\big) \end{aligned} \tag{35}$$

and

$$\begin{aligned} c^{(2)}_{\vec{k}\sigma\vec{k}'\lambda}(\infty) &= \lim_{s\to 0}[sy^{(2)}_{c\vec{k}\sigma\vec{k}'\lambda}(s)] = \lim_{s\to 0}\{sY^{(2)}_{c\vec{k}\sigma\vec{k}'\lambda}[s-i(\omega_{c\sigma\lambda}-\omega_0)]\} \\ &= (H_{c\vec{k}\sigma\vec{k}'\lambda;a\vec{k}\sigma}H_{a\vec{k}\sigma;0}/\hbar^2)(\omega_\lambda-\omega_{ac}+i\tfrac{1}{2}\gamma)^{-1} \\ &\quad\times\big((\omega_\lambda-\omega_\sigma+i\tfrac{1}{2}\Gamma)^{-1}+|V_{ab}/\hbar|^2\{[\omega_\lambda-\omega_\sigma+i(2\epsilon+\tfrac{1}{2}\Gamma)](\omega_\lambda-\omega_{bc}+i\epsilon) \\ &\quad\times[\omega_\lambda-\omega_{ac}+i(2\epsilon+\tfrac{1}{2}\gamma)]\}^{-1}+|V_{ab}/\hbar|^4 \\ &\quad\times\{[\omega_\lambda-\omega_\sigma+i(4\epsilon+\tfrac{1}{2}\Gamma)][\omega_\lambda-\omega_{ac}+i(2\epsilon+\tfrac{1}{2}\gamma)](\omega_\lambda-\omega_{bc}+i\epsilon)(\omega_\lambda-\omega_{bc}+i3\epsilon) \\ &\quad\times[\omega_\lambda-\omega_{ac}+i(4\epsilon+\tfrac{1}{2}\gamma)]\}^{-1}\big) , \end{aligned} \tag{36}$$

where

$$\omega_{ba}=\omega_b-\omega_a , \quad \omega_{ac}=\omega_a-\omega_c , \quad \text{and } \omega_{bc}=\omega_b-\omega_c . \tag{37}$$

The other probability amplitudes $c_0(t)$ and $a_{\vec{k}\sigma}(t)$ vanish as $t\to\infty$.

Now the probability P_b of the atom being in the excited state $|b\rangle$ at $t\to\infty$, after having absorbed the incident radiation, is obtained by summing the probability $|b_{\vec{k}\sigma}(\infty)|^2$ over the frequency and polarization of the incident radiation. The integrals are evaluated by contour integration. The result is

$$\begin{aligned} P_b = &(V^2/\hbar^2)\Gamma\{(\Gamma+2\epsilon)[\omega^2_{ba}+(\epsilon+\tfrac{1}{2}\gamma)^2]\}^{-1} \\ &\times\big(1-(V^2/\hbar^2\epsilon)(\Gamma+2\epsilon)(3\epsilon+\tfrac{1}{2}\gamma)\{(\Gamma+4\epsilon)[\omega^2_{ba}+(3\epsilon+\tfrac{1}{2}\gamma)^2]\}^{-1} \\ &+(V^4/8\hbar^4\epsilon^2)(\Gamma+2\epsilon)(\gamma+8\epsilon)(\gamma+10\epsilon)\{(\Gamma+6\epsilon)[\omega^2_{ba}+(3\epsilon+\tfrac{1}{2}\gamma)^2][\omega^2_{ba}+(5\epsilon+\tfrac{1}{2}\gamma)^2]\}^{-1}\big) , \end{aligned} \tag{38}$$

where V_{ab} has been assumed to be real and replaced by V.

The above probability depends on the coupling perturbation V, the decay constant ϵ of the perturbing field, and on the frequency difference ω_{ab} between the excited states. It is evident from Eq. (38) that the probability is maximum when the two excited states are degenerate (i.e., $\omega_a = \omega_b$). The expression for P_b in Eq. (38) is a second-order solution which converges very rapidly for small perturbations such that $V/\hbar\epsilon$ is less than unity. When $\epsilon \to 0$, Eq. (38) is valid only if $V/\hbar\epsilon < 1$. However, an exact solution can be obtained for $\epsilon = 0$ and $V \neq 0$ (see Ref. 4) which yields $P_b = 0$. Further discussion on P_b is presented in Sec. IV.

The probability P_c of the atom being in the ground state $|c\rangle$ after having absorbed the incident radiation and having emitted a photon is obtained by summing the probability $|c_{k\sigma k'\lambda}(\infty)|^2$ over the frequency and polarization of the incident radiation and over frequency, direction, and polarization of the emitted radiation. This probability[12] is also obtained very simply by the normalization condition and therefore it can be written as

$$P_c = 1 - P_b \ . \tag{39}$$

Thus we see that by using a perturbing potential of the form defined in Eq. (1) we are able to "trap" some energy from the incident radiation with the excited atoms in the metastable states. The next step is to release the "trapped" energy whenever it is required. This process is discussed in Sec. III.

III. ENERGY-RELEASE MECHANISM

Here we discuss the "release" mechanism of the trapped energy with the atoms in their metastable states. This could easily be achieved by applying a static field[3] which couples the metastable state with the decaying one and causes the atom to decay to the ground state through emission of radiation. Thus the trapped energy is released in the form of a radiation pulse.

In fact, this problem has already been studied by Fontana and Lynch.[13] However, we shall present here the important steps of their calculations. The only difference between the present problem and theirs is in the initial probability of the metastable state. Fontana and Lynch assume the above probability to be unity whereas in our case it is P_b [see Sec. II, Eq. (38)]. Our main interest here is to find the frequency and angular distribution of the emitted radiation. For this, we have to solve again the Schrödinger equation of motion for the system.

The total Hamiltonian for the system is

$$\mathcal{H} = H_0 + H + V' \tag{40}$$

where H_0 is the unperturbed Hamiltonian for the atom and the radiation field, H represents the interaction of the radiation with the atom as defined in Sec. II, and V' is the static external coupling field.

The following states represent the eigenvectors of the unperturbed Hamiltonian H_0:

$$|a;0\rangle \equiv |a\rangle\,|0\rangle_{\rm rad}\,, \quad |b;0\rangle \equiv |b\rangle\,|0\rangle_{\rm rad}\,,$$

and

$$|c;\vec{k}\lambda\rangle \equiv |c\rangle\,|\vec{k}\lambda\rangle \tag{41}$$

with the corresponding eigenfrequencies ω_a, ω_b, and $\omega_{c\lambda}$, where

$$\omega_{c\lambda} = \omega_c + \omega_\lambda \tag{42}$$

and $|0\rangle_{\rm rad}$ and $|\vec{k}\lambda\rangle_{\rm rad}$ represent the states of the radiation field with no photons present and a photon being emitted with wave vector $\vec{k}$, frequency ω_λ, and polarization λ, respectively.

The state vector of the system at any time t can be written:

$$|\psi(t)\rangle = a_0(t)\,|a;0\rangle\, e^{-i\omega_a t} + b_0(t)\,|b;0\rangle\, e^{-i\omega_b t} + \sum_{\vec{k}\lambda} c_{\vec{k}\lambda}(t)\,|c;\vec{k}\lambda\rangle\, e^{-i\omega_{c\lambda} t}\ , \tag{43}$$

where $a_0(t)$, $b_0(t)$, and $c_{\vec{k}\lambda}(t)$ are the probability amplitudes in the corresponding states.

The Schrödinger equation yields the following equations for the probability amplitudes.

$$i\hbar\dot{a}_0(t) = \sum_{\vec{k}\lambda} H_{a0;c\vec{k}\lambda}\, c_{\vec{k}\lambda}(t) \exp[i(\omega_a - \omega_{c\lambda})t] + V'_{ab}\, b_0(t)\exp[i(\omega_a - \omega_b)t]\,, \tag{44}$$

$$i\hbar\dot{b}_0(t) = V'_{ba}\, a_0(t)\exp[i(\omega_b - \omega_a)t]\,, \tag{45}$$

$$i\hbar\dot{c}_{\vec{k}\lambda}(t) = H_{c\vec{k}\lambda;a0}\, a_0(t)\exp[i(\omega_{c\lambda} - \omega_a)t]\,, \tag{46}$$

where

$$H_{a0;c\vec{k}\lambda} = \langle a;0\,|H|\,c;\vec{k}\lambda\rangle\,, \qquad V'_{ab} = \langle a|V'|b\rangle\,. \tag{47}$$

Here again it is assumed that H does not couple the states $|a\rangle$ and $|b\rangle$ (i.e., no radiation transition between $|a\rangle$ and $|b\rangle$ is assumed) and also the external perturbation V' does not mix the ground state with the excited states. The diagonal matrix elements of V' are considered to be zero in Eqs. (44)–(46), though the case of nonvanishing diagonal matrix elements of V' can be treated easily as described in Sec. II.

By making the following substitutions for the amplitudes:

$$A_n = a_n \exp[i(\omega_a - \omega_n)t]\,, \tag{48}$$

where a_n stands for the original amplitudes, Eqs. (44)–(46) reduce to

$$i\hbar\dot{A}_0(t)=\sum_{\vec{k}\lambda} H_{a0;c\vec{k}\lambda}C_{\vec{k}\lambda}(t)+V'_{ab}B_0(t)\,, \qquad (49)$$

$$i\hbar[\dot{B}_0(t)+i(\omega_b-\omega_a)B_0(t)]=V'_{ba}A_0(t)\,, \qquad (50)$$

$$i\hbar[\dot{C}_{\vec{k}\lambda}(t)+i(\omega_{c\lambda}-\omega_a)C_{\vec{k}\lambda}(t)]=H_{c\vec{k}\lambda;a0}A_0(t)\,. \qquad (51)$$

Now, Laplace transforming the above equations and using the initial condition

$$b_0(0)=P_b^{1/2},\quad a_0(0)=c_{\vec{k}\lambda}(0)=0\,, \qquad (52)$$

one obtains

$$[s+\tfrac{1}{2}\gamma]\,Y_{a0}(s)=-i(V'_{ab}/\hbar)Y_{b0}(s)\,, \qquad (53)$$

$$[s+i(\omega_b-\omega_a)]\,Y_{b0}(s)=P_b^{1/2}-i(V'_{ba}/\hbar)Y_{a0}(s)\,, \qquad (54)$$

$$[s+i(\omega_{c\lambda}-\omega_a)]\,Y_{c\vec{k}\lambda}(s)=-i(H_{c\vec{k}\lambda;a0}/\hbar)Y_{a0}(s)\,, \qquad (55)$$

where the $Y_n(s)$'s are the Laplace transforms of the amplitudes (A_n's) in Eq. (48). The constant γ in Eq. (53) is, in fact, a function of s and defined by a relation similar to that of Eq. (27). However, using the same argument as discussed earlier in Sec. II, we can consider γ to be constant representing the decay constant of the unperturbed state $|a\rangle$.

Equations (53)–(55) can, now, be solved exactly, yielding the results

$$Y_{a0}(s)=-i(V'_{ab}/\hbar)P_b^{1/2}\times\{(s+\tfrac{1}{2}\gamma)[s+i(\omega_b-\omega_a)]+|V'_{ba}/\hbar|^2\}^{-1}\,, \qquad (56)$$

$$Y_{b0}(s)=[s+\tfrac{1}{2}\gamma]P_b^{1/2}\times\{(s+\tfrac{1}{2}\gamma)[s+i(\omega_b-\omega_a)]+|V'_{ba}/\hbar|^2\}^{-1}\,, \qquad (57)$$

$$Y_{c\vec{k}\lambda}(s)=-(H_{c\vec{k}\lambda;a0}V'_{ab}/\hbar^2)P_b^{1/2}\times([s+i(\omega_{c\lambda}-\omega_a)]\{(s+\tfrac{1}{2}\gamma)[s+i(\omega_b-\omega_a)]+|V'_{ba}/\hbar|^2\})^{-1}\,. \qquad (58)$$

Using the inverse Laplace transforms of the above $Y(s)$'s, one can determine the time-dependent amplitudes. However, since we are interested only in the frequency and angular distribution of the emitted radiation, we find the amplitudes at $t\rightarrow\infty$. Thus, using again the final-value theorem,[11] we obtain

$$c_{\vec{k}\lambda}(\infty)=(H_{c\vec{k}\lambda;a0}V'_{ab}/\hbar^2)P_b^{1/2}\times[(\omega_\lambda-\omega_{ac}+i\tfrac{1}{2}\gamma)(\omega_\lambda-\omega_{bc})-|V'_{ab}/\hbar|^2]^{-1}\,, \qquad (59)$$

where ω_{ac} and ω_{bc} are defined in Eq. (37). The other amplitudes vanish at $t\rightarrow\infty$.

The probability of the atom being in the ground state $|c\rangle$ after having emitted a photon of wave vector $\vec{k}$, frequency ω_λ, and polarization λ is given by

$$|c_{\vec{k}\lambda}(\infty)|^2=|H_{c\vec{k}\lambda;a0}/\hbar|^2|V'_{ab}/\hbar|^2P_b\times\{[(\omega_\lambda-\omega_{ac})(\omega_\lambda-\omega_{bc})-|V'_{ab}/\hbar|^2]^2+(\omega_\lambda-\omega_{bc})^2\tfrac{1}{4}\gamma^2\}^{-1}\,. \qquad (60)$$

This result is the same as Eq. (27) of Ref. 13, except for a factor of P_b. The angular distribution of the emitted radiation would depend on the factor $|H_{c\vec{k}\lambda;a0}|^2$ which in turn depends on the interaction Hamiltonian H. In the absence of multiphoton process, it contains only $\vec{p}\cdot\vec{A}$ terms as mentioned earlier. When the state $|a\rangle$ is connected to the state $|c\rangle$ through an electric-dipole transition, the angular distribution of the emitted radiation depends on $|\langle a|\vec{r}|c\rangle\cdot\hat{e}_\lambda|^2$, where $\vec{r}$ is the position vector of the electron in the atom and $\hat{e}_\lambda$ is the direction of polarization of the emitted photon. The total emitted energy E per atom is obtained by summing the probability $|c_{\vec{k}\lambda}(\omega)|^2$ in Eq. (60) over frequency, direction, and polarization of the emitted photon:

$$E=\hbar\bar{\omega}\sum_{\vec{k}\lambda}|c_{\vec{k}\lambda}(\infty)|^2\,, \qquad (61)$$

i.e.,

$$E=\hbar\bar{\omega}P_b\,, \qquad (62)$$

where $\bar{\omega}$ represents the average frequency of the photon. The summation over $\vec{k}$ in Eq. (61) is replaced by integrations over frequency ω_λ and the direction of emission. The integral over frequency is evaluated through contour integration.

In the presence of large numbers of atoms and in the absence of a quantization field (such as a magnetic field) the emitted radiation has equal probability in all directions. Thus the energy released by the atoms per unit volume per unit solid angle is given by

$$nE/4\pi=n\hbar\bar{\omega}P_b/4\pi\,, \qquad (63)$$

where n is the density of the gas.

In the present problem, we have completely ignored the collision effects even though collision effects are quite prominent at moderate pressures. However, one can reduce the effects by reducing the pressure of the gas to a minimum and still "trap" a significant amount of energy. This will be further discussed in Sec. IV with some specific examples.

IV. DISCUSSION

The variation of the probability P_b [Eq. (38)] with the decay constant ϵ is shown in Fig. 1 for

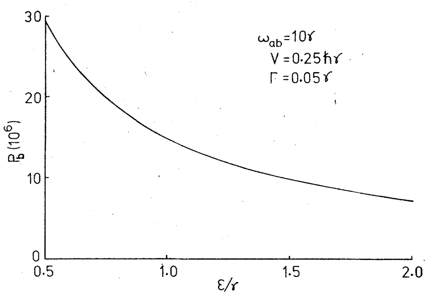

FIG. 1. Variation of P_b with ϵ for constant V.

$\omega_{ab}=10\gamma$ and $\Gamma=0.05\gamma$ [Γ is very small compared to γ for not very strong radiation (see Ref. 7)]. The value of V is chosen to be $0.25\hbar\gamma$ which is always less than $\hbar\epsilon$ used in the graph [the convergence condition for Eq. (38) is $V<\hbar\epsilon$]. It is observed that the probability decreases monotonically with the increase in ϵ.

Figure 2 shows again the variation of the probability P_b with ϵ but this time the coupling potential V is also varied so that the convergence condition is satisfied ($V=0.5\hbar\epsilon$ and $0.35\hbar\epsilon$). It is interesting to note that a maximum exists in P_b for a certain value of V and ϵ. The decrease in the probability with decreasing ϵ is due to the fact that the coupling field stays for longer time and thus the nondecaying state $|b\rangle$ gets more time to decay via the state $|a\rangle$. Similarly, at large ϵ, the probability decreases because the coupling field is only present for a very short time. During this time not much probability is transferred by the coupling field to the state $|b\rangle$ from the state $|a\rangle$ in the absorption process. An increase in V for the same ϵ increases the probability as shown in Fig. 2. These results suggest that suitable values for V and ϵ can be chosen to obtain a maximum probability for trapping the atom in the nondecaying state. In the case of two degenerate upper levels, the probabilities in Figs. 1 and 2 would increase by a factor of 100 for the same V and ϵ $\{P_b \propto [\omega_{ab}^2+(\frac{1}{2}\gamma+\epsilon)^2]^{-1}$; see Eq. (38)$\}$.

The energy released by the atoms per unit volume per unit solid angle is $n\hbar\bar{\omega}P_b/4\pi$ as mentioned earlier. This energy is quite significant and moreover it is emitted in a very short time; thus the power produced by the device would be quite large. For example, if H atoms were used in the device where the metastable state $2\,^2S_{1/2}$ and the decaying state $2\,^2P_{1/2}$ are coupled by an electric field (the other higher states are assumed to be barred from the absorption of radiation[2]), the energy released per unit solid angle by 1 cm^3 of gas would be ~1 erg/cm^3 at a pressure of 0.001 atm and temperature of 300 °K for $V=0.25\hbar\gamma$, $\epsilon=0.5\gamma$, and $\Gamma=0.05\gamma$. The corresponding power would be ~100 W/cm^3. Thus, it is apparent that if such a device is constructed, it would have numerous applications. However, there is a practical difficulty of finding such a system with an infinitely long-lived metastable state. In fact, in the absence of collisions, the hydrogen atom chosen in the above example has $\frac{1}{7}$-sec lifetime[14] in the $2\,^2S_{1/2}$ state due to the two-photon decay to the ground state ($1\,^2S_{1/2}$). The collisional decay of the $2\,^2S_{1/2}$ state of H atom is also quite significant [$10^{-8}n$ atoms/sec (Ref. 14)]. Therefore, H atom would not be a very suitable candidate for a device to be used for storing energy. An extensive investigation is desired to look for suitable candidates.

Besides using the device for storing energy, one might speculate that the effect may be used in constructing "solar lasers." Here, we *do not* really need the lifetime of the metastable state to be infinitely large. For this, one has to consider a four-level system: two lower and two upper states with one of the upper states being a metastable one. The system is excited to the metastable state through the energy "trapping" mechanism by absorbing the solar radiation. For a suitable energy difference between the ground state and the other lower state, population inversion between the lower state (other than ground state) and the upper (decaying) state may be achieved at the triggering time. This would cause the system to lase. Molecular systems seem to be more suitable for constructing such "solar lasers," where the higher vibrational states of the ground electronic state may serve as the fourth state for the laser action.

Another possible application may be in constructing "solar lamps" (which would emit monochromatic radiation), where the lifetime of the metastable state, again, does not have to be infinitely large. For this, a continuous flow of the gas (three-level atomic gas which has trapped the solar energy by the mechanism described here) has to be maintained through a triggering zone where the emission of radiation is desired. The only limitation would be that the time between the events, when

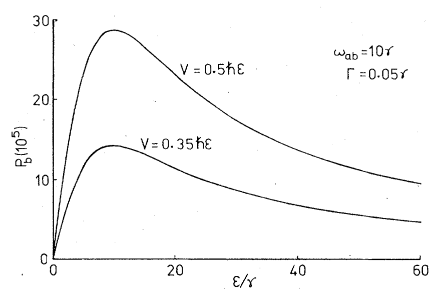

FIG. 2. Variation of P_b with ϵ for varying V.

the solar radiation is trapped and when it is released (in the triggering zone), has to be less than the lifetime of the metastable state.

Although the above discussion has been presented on the basis of a hypothetical model, it is quite encouraging to see that there are many atomic and molecular systems (e.g., H, He, Be, N, O, Hg, I_2, O_2, etc.; see Refs. 15 and 16) which might behave like the one described here. The next task which ought to be pursued is to investigate these systems in detail.

ACKNOWLEDGMENT

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[1]R. P. Srivastava, Phys. Lett. 62A, 475 (1977).

[2]The three-level atomic gas can be produced out of normal atoms by using a proper filter in the incident radiation. This filter bars the unwanted levels from participating in the absorption process. In fact, the device is not limited to atomic systems only; rather it can consist of any three-level quantum system such as atomic, molecular, or nuclear systems.

[3]The external coupling field may be an electric field or a magnetic field, depending on the states to be coupled. For example, in the case of H, $2\,^2S_{1/2}$ (metastable) and $2\,^2P_{1/2}$ states can be coupled by an electric field.

[4]R. P. Srivastava and P. R. Fontana, J. Phys. B 7, 911 (1974).

[5]Choice of an exponentially decaying field is mainly due to the theoretical simplicity. For experimental work, any short pulse which shuts off before completion of the resonance fluorescence process will serve the purpose.

[6]G. Baym, *Lectures on Quantum Mechanics* (Benjamin, New York, 1969).

[7]W. Heitler, *The Quantum Theory of Radiation*, 3rd ed. (Oxford U. P., London, 1960).

[8]J. M. Schurr, Int. J. Quantum Chem. 5, 13 (1971).

[9]P. R. Fontana and P. Thomann, Phys. Rev. A 13, 1512 (1976).

[10]For continuous radiation with energy $I_0(\omega_\sigma)\,d\omega_\sigma$ (energy per unit area per second) incident from a given direction within solid angle $d\Omega$, Γ can be expressed in terms of $I_0(\omega_{ac})$ as in Eq. (15) on page 200 of Ref. 7. Here the intensity $I_0(\omega_\sigma)$ is assumed to be constant over the frequency range $(\omega_a - \omega_b)$ of interest.

[11]D. K. Cheng, *Analysis of Linear Systems* (Addison-Wesley, Reading, Mass., 1961).

[12]The probability P_c obtained from $|c_{\vec{k}\sigma\vec{k}'\lambda}(\infty)|^2$ by summing over the incident and emitted photons states agrees with that obtained using Eq. (39) as verified by the author up to $V^2/\hbar^2$ terms in P_c. Beyond $V^2/\hbar^2$ terms, the integrals become very cumbersome and it is not necessary to go that route.

[13]P. R. Fontana and D. J. Lynch, Phys. Rev. A 2, 347 (1970).

[14]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-electron Atoms* (Springer-Verlag, Berlin, 1957).

[15]*Atomic Energy Levels*, U. S. Natl. Bur. Stand., edited by C. A. Moore (U. S. GPO, Washington, D.C.), Vol. I (1949), Vol. II (1952), and Vol. III (1958).

[16]G. Herzberg, *Spectra of Diatomic Molecules* (Van Nostrand Company, New York, 1950).