

Absorption of a pulse by an optically dense medium: An argument for field quantization

P. R. Berman

*Department of Physics and Michigan Center for Theoretical Physics, University of Michigan,
450 Church Street, Ann Arbor, Michigan 48109-1040*

J.-L. Le Gouët

*Laboratoire Aimé Cotton, CNRS UPR3321, University Paris Sud, Bâtiment 505, Campus Universitaire,
91405 Orsay, France*

(Received 18 August 2010; accepted 27 December 2010)

The theory of the absorption of a weak optical pulse by an optically dense medium is shown to lead to unphysical results unless the radiation field is quantized. In contrast to the photoelectric effect, the atom-field dynamics for pulsed field absorption can be obtained using elementary quantum mechanics without imposing any assumptions on the nature of the detection process. As such, pulsed field absorption offers distinct advantages over the photoelectric effect as a proving ground for field quantization. If the classical field pulse is replaced by a quantized, multimode field state, many classical field results are recovered without the inconsistencies that arise in the classical field calculation. © 2011 American Association of Physics Teachers.
[DOI: 10.1119/1.3549235]

I. INTRODUCTION

Processes such as the photoelectric effect and Compton scattering are given as textbook examples of the need to quantize the radiation field in free space, that is, evidence for photons. However, calculations of both the photoelectric effect and the Compton scattering can be carried out using classical excitation fields.¹ Lamb and Scully² showed that without resorting to field quantization, all the features of the photoelectric effect explained by Einstein in his 1905 paper³ can be recovered. Thus, the justification given for field quantization in most textbooks is not convincing.

The Lamb–Scully calculation is correct as far as it goes. However, there are certain limits for both the photoelectric effect and the Compton scattering that cannot be explained consistently using pulsed classical radiation fields. For the photoelectric effect, there are problems when the energy in the pulse is less than or on the order of the work function of the metal. In the Compton effect, there are problems with the classical field description when the pulse energy is equal to the change in energy undergone by the electron in the scattering process.

To show that the assumption of classical fields leads to erroneous results, it is necessary to include changes in the classical field pulse resulting from interaction with matter. Unfortunately, such a calculation is not easy for the photoelectric effect and Compton scattering. However, if we consider absorption of a classical pulse by an optically dense medium, it is relatively easy to account for the field dynamics using Maxwell's equations. The field is simply absorbed as it propagates in the medium. The interaction of the classical field pulse with the atoms in the ensemble is treated by elementary quantum mechanics. With such a model, we can understand quickly that field quantization is needed for a consistent picture of matter-field interactions.

Imagine that the medium consists of an ensemble of two-level atoms with transition frequency ω_0 . A classical field pulse with energy $\hbar\omega_0$ is incident on the medium and is totally absorbed. We will show that despite the fact that the pulse is totally absorbed, there is a probability of $1/e$ that all

the atoms remain in their ground states, which is a paradoxical result. Moreover, there is a nonvanishing probability to have more than one atom excited by the pulse, a process that violates energy conservation. This simple example shows that a classical field description fails if the pulse energy is comparable to a relevant quantum transition frequency. We argue that the interaction of any classical field pulse with a two-level quantum system leads to inconsistencies if the field is not quantized.

Our goal is to provide a calculation of field absorption, accessible to advanced undergraduate and beginning graduate students, that demonstrates the need for field quantization. We show that all the features of the classical field calculation are reproduced if the classical field is replaced by a multimode quantum coherent state, but none of the inconsistencies present in the classical field calculation persist when the quantized coherent state field is used. This calculation represents a more convincing argument for the need to quantize the field than do discussions of the photoelectric effect and Compton scattering.

As with the photoelectric effect and Compton scattering, our argument relies on the quantized nature of matter. From an operational point of view, it seems to be necessary to look at the interaction of the radiation field with a quantized material system to show that it is necessary to quantize the field.

II. CLASSICAL PULSE ABSORPTION

We consider the absorption of a classical optical pulse by an optically dense medium. The field evolution is described by Maxwell's equations and the atom-field interaction is treated quantum mechanically. The polarization of the medium that leads to field absorption is calculated as the expectation value of the dipole moment of the atoms, as in standard semiclassical theories involving the Maxwell–Bloch equations. The problem of a classical pulse propagating in a medium consisting of an ensemble of two-level atoms is usually complicated due to pulse distortion and reshaping as the pulse propagates in the medium. To simplify the calculation, we adopt a model in which, although optically dense, the

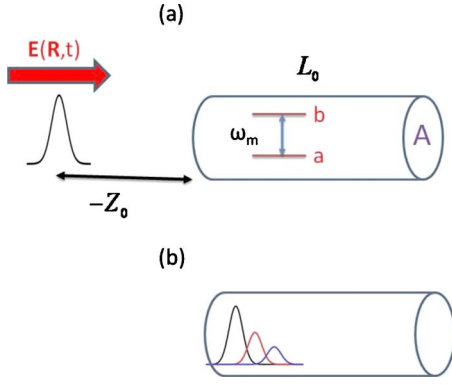


Fig. 1. (a) A classical field pulse is incident on a medium of inhomogeneously broadened atoms. (b) The field enters the medium and propagates with speed c and without distortion, but is exponentially damped in the medium.

medium has an effective index of refraction equal to unity for the incident pulse, and the pulse propagates without shape distortion due to inhomogeneous broadening. The only effect of the medium is to result in an exponential decrease in the pulse intensity as it propagates in the medium.

To arrive at this simplified situation, we make a number of assumptions that do not compromise the underlying physics of interest in this paper. The medium consists of an inhomogeneously broadened ensemble of stationary, two-level atoms located uniformly in a cylinder with cross-sectional area A and length L_0 [see Fig. 1(a)], located between $Z=0$ and $Z=L_0$. The ground state of atom m is denoted by $|a_m\rangle$, its excited state by $|b_m\rangle$, and its transition frequency by ω_m . Due to inhomogeneous broadening, the atoms have a distribution of atomic transition frequencies centered about the central frequency ω_0 . The detuning of ω_m from the central frequency ω_0 is denoted by

$$\delta_m = \omega_m - \omega_0. \quad (1)$$

It is assumed that the distribution of δ_m is governed by

$$F(\delta_m) = \frac{1}{\sqrt{\Delta}} e^{-\delta_m^2/\Delta^2}, \quad (2)$$

where Δ is a measure of the inhomogeneity of the sample. The excited state decay rate of the atoms is equal to γ .

A classical pulse is incident from the left. The pulse, which has a cross-sectional area A to match the sample cross-sectional area, is polarized in the x direction and propagates in the z direction. The pulse is transform-limited with a temporal pulse width τ . It is assumed that the spectral width of the pulse, $\delta\omega = \tau^{-1}$, is much less than Δ , ensuring that each frequency component of the field is absorbed equally by the sample; the pulse duration is much less than the excited state lifetime of the atoms and any transverse relaxation rate, allowing us to neglect any decay as the pulse excites an atom; and the energy in the pulse is equal to $\hbar\omega_0$, the average excitation energy of an atom in the sample.

We choose the energy in the pulse as $\hbar\omega_0$ because it allows us to see in a dramatic fashion the unphysical results that come from the assumption of a classical field pulse. However, any field energy would work equally well. The field pulse is not a single photon pulse because we are considering a classical field. It is a pulse with a well-defined total energy $\hbar\omega_0$. For a classical field, any energy is allowed.

The medium is taken to be optically dense so that $\alpha L_0 \gg 1$, where α is the absorption coefficient of the medium. To neglect the effects of atomic fluctuations, we assume that the number of atoms in a slice of the medium of length $dZ = \lambda_0 = 2\pi\omega_0/c$ is much greater than unity, that is,

$$\rho A \lambda_0 \gg 1, \quad (3)$$

where ρ is the atomic density. Equation (3) is the validity condition for the use of Maxwell's equations for the field variables that are macroscopically averaged over a slice of the medium with a thickness that is much less than a wavelength so that in such a slice the spatial phase of the field is the same for all the atoms. In contrast, we assume that

$$\rho \lambda_0^3 \ll 1 \quad (4)$$

to guarantee that the index of refraction of the medium is approximately equal to unity and atom-atom interactions can be neglected.⁴ Equations (3) and (4) imply that $A \gg \lambda_0^2$, allowing us to neglect diffraction of the pulse.

The result of all these assumptions is that the pulse enters the medium without reflection and propagates in the medium with exponentially decreasing amplitude but without shape distortion [see Fig. 1(b)]. We assume that the pulse travels at most a few pulse widths in the medium before it is absorbed and consider only times sufficiently short to ensure that the atoms do not decay, but sufficiently long to guarantee that the pulse has been totally absorbed. In other words, we consider only times for which the optical pulse has transferred all its energy to the internal energy of the atoms.

As we have stressed, all the assumptions of the medium and the incident pulse are adopted to simplify the calculations. The same inconsistencies that we will find would arise even if these assumptions were relaxed. There are physical systems that satisfy all our assumptions. For example, if picosecond pulses are sent into a medium whose active atoms consist of impurity ions embedded in a host material,⁵ all our assumptions can be realized.

The initial electric field is taken as

$$E(Z,0) = \frac{1}{2} \hat{x} E_0 e^{ik_0 Z} f(Z+Z_0) + \text{complex conjugate}, \quad (5)$$

where $f(Z)$ is a smooth, positive envelope function centered at $Z=0$ with width $(\delta Z) \ll Z_0$ ($Z_0 > 0$) and E_0 (assumed real) is the pulse amplitude. It is assumed that the amplitude is constant over the cross-sectional area A of the medium and zero outside this range. The initial pulse is centered at $Z = -Z_0 < 0$. The pulse is quasimonochromatic, $k_0(\delta Z) \gg 1$, and the parameter k_0 serves as the average wave number.

For definiteness, we take the envelope function to be

$$f(Z) = e^{-Z^2/2(\delta Z)^2}. \quad (6)$$

The energy in the initial pulse, W_0 , is set equal to $\hbar\omega_0$; that is,

$$W_0 = \frac{1}{2} \epsilon_0 E_0^2 A \int_{-\infty}^{\infty} f^2(Z) dZ = \frac{1}{2} \sqrt{\pi} \epsilon_0 E_0^2 A (\delta Z) = \hbar\omega_0, \quad (7)$$

where ϵ_0 is the vacuum permittivity. We solve for E_0 in Eq. (7) and find

$$E_0 = \left(\frac{2\hbar\omega_0}{\epsilon_0 A (\delta Z) \sqrt{\pi}} \right)^{1/2}. \quad (8)$$

Once the pulse enters the medium, it decays exponentially. That is, inside the medium the pulse electric field is given by

$$E(Z, t) = \frac{1}{2} \hat{\mathbf{x}} E_0 e^{i(k_0 Z - \omega_0 t)} e^{-\alpha Z/2} f(Z + Z_0 - ct) + \text{complex conjugate}, \quad (9)$$

where $\omega_0 = k_0 c$ and α is the absorption coefficient. For an inhomogeneously broadened medium having a distribution of frequencies given by Eq. (2) and for a pulse bandwidth much less than the inhomogeneous width, $c/(\delta Z) = \tau^{-1} \ll \Delta$, the absorption coefficient is equal to⁶

$$\alpha = \frac{\sqrt{\pi} \rho \omega_0 \mu^2}{\hbar \epsilon_0 c \Delta}, \quad (10)$$

where μ (assumed real) is the x component of the dipole matrix element between the ground and the excited states of an atom.

As the pulse passes atom m located at $Z = Z_m$, it excites that atom. The atom-field interaction for this atom is taken to be $-\mu E(Z_m, t)$. As a consequence, it follows from Schrödinger's equation that the evolution of the excited state amplitude $b(Z_m, \delta_m, t)$ for atom m (in the interaction representation and in the rotating wave approximation) is governed by

$$\dot{b}(Z_m, \delta_m, t) = i[\mu E(Z_m, t)/2\hbar] a(Z_m, \delta_m, t), \quad (11)$$

where $a(Z_m, \delta_m, t)$ is the ground state amplitude of atom m . The assumption that the pulse area is much less than unity allows us to calculate the excited state amplitude using perturbation theory, that is, by setting $a(Z_m, \delta_m, t) \approx 1$. With this assumption and the use of Eqs. (6) and (9), we find that the excited state amplitude $b(Z_m, \delta_m)$ after the passage of the pulse is

$$\begin{aligned} b(Z_m, \delta_m) &= -i\chi e^{-\alpha Z_m/2} e^{ik_0 Z_m} \int_{-\infty}^{\infty} dt e^{-t^2/2\tau^2} e^{i\delta_m t} \\ &= -i\sqrt{2\pi}\chi\tau e^{-\alpha Z_m/2} e^{ik_0 Z_m} e^{-\delta_m^2 \tau^2/2}, \end{aligned} \quad (12)$$

where $\chi = -\mu E_0/2\hbar$ is one-half the atom-field Rabi frequency.

The resultant final state wave vector for the atoms in the sample once the pulse is totally absorbed is

$$\begin{aligned} &|\psi(Z_1, Z_2, \dots, Z_N; \delta_1, \delta_2, \dots, \delta_N)\rangle \\ &= \prod_{m=1}^N [a(Z_m, \delta_m)|a_m\rangle + b(Z_m, \delta_m)|b_m\rangle e^{-i\omega_m t}], \end{aligned} \quad (13)$$

where $N = \rho A L_0$ is the total number of atoms in the sample and $a(Z_m, \delta_m)$ is the ground state amplitude for atom m once the pulse has passed. From probability conservation, it follows that

$$|a(Z_m, \delta_m)|^2 = 1 - |b(Z_m, \delta_m)|^2. \quad (14)$$

The state vector (13) is a product state and there is no entanglement. The average total energy in the sample, W_f , after the pulse has been absorbed is

$$\begin{aligned} W_f &= \sum_{m=1}^N \hbar \omega_m |b(Z_m, \delta_m)|^2 \\ &\rightarrow \hbar \rho A \int_0^{L_0} dZ \int_{-\infty}^{\infty} d\delta F(\delta) (\delta + \omega_0) |b(Z, \delta)|^2, \end{aligned} \quad (15)$$

where the discrete sum has been converted to integrals over space and frequency.

If we combine Eqs. (2), (12), and (15), we find

$$\begin{aligned} W_f &= \frac{2\pi(\chi\tau)^2 \hbar \rho A}{\sqrt{\pi}\delta_0} \int_0^{L_0} dZ \int_{-\infty}^{\infty} d\delta (\delta + \omega_0) e^{-\alpha Z} e^{-\delta^2 \tau^2} e^{-\delta^2/\delta_0^2} \\ &= \frac{2\pi\chi^2 \tau \hbar \rho A \omega_0}{\alpha} = \frac{\sqrt{\pi}\epsilon_0 A (\delta Z) E_0^2}{2}, \end{aligned} \quad (16)$$

where the inequalities $\alpha L_0 \gg 1$ and $\Delta \gg \tau^{-1}$ have been used. With E_0 given by Eq. (8), it follows that

$$W_f = \hbar \omega_0, \quad (18)$$

consistent with conservation of energy.

Although the average energy in the sample is $\hbar \omega_0$, there is a probability that either no atom is excited or more than one atom is excited. The probability for no atom to be excited is

$$P_0 = \prod_{m=1}^N |a(Z_m, \delta_m)|^2 = \prod_{m=1}^N [1 - |b(Z_m, \delta_m)|^2] \quad (19)$$

$$\begin{aligned} &= 1 - \sum_{m=1}^N |b(Z_m, \delta_m)|^2 \\ &+ \frac{1}{2!} \sum_{m, m'=1}^N |b(Z_m, \delta_m)|^2 |b(Z_{m'}, \delta_{m'})|^2 \epsilon_{m, m'} \end{aligned} \quad (20)$$

$$\begin{aligned} &- \frac{1}{3!} \sum_{m, m', m''=1}^N |b(Z_m, \delta_m)|^2 |b(Z_{m'}, \delta_{m'})|^2 \\ &\times |b(Z_{m''}, \delta_{m''})|^2 \epsilon_{m, m', m''} + \dots, \end{aligned} \quad (21)$$

where $\epsilon_{i, j, k, \dots}$ vanishes if any of its two indices are equal and is equal to unity otherwise. In the limit in Eq. (3), we can set $\epsilon_{i, j, k, \dots}$ equal to unity and replace the sums in Eq. (21) by integrals of the type given in Eq. (15). In this way, we find

$$P_0 = 1 - S + \frac{S^2}{2!} - \frac{S^3}{3!} + \dots = e^{-S}, \quad (22)$$

where

$$S = \hbar \rho A \int_0^{L_0} dZ \int_{-\infty}^{\infty} d\delta F(\delta) |b(Z, \delta)|^2 = 1. \quad (23)$$

Thus, we find that the probability that no atom is excited by the pulse is equal to

$$P_0 = 1/e. \quad (24)$$

This result is nonphysical because if no atom is excited, the pulse cannot be absorbed. Following a similar procedure, we can show that the probability P_n that exactly n atoms are excited (and $N-n$ remain in their ground states) is given by

$$P_n = C_n^N \left(1 - \frac{1}{N}\right)^{N-n} \left(\frac{1}{N}\right)^n \quad (25)$$

$$= \frac{N(N-1)\dots(N-n+1)}{n!} \left(1 - \frac{1}{N}\right)^{N-n} \left(\frac{1}{N}\right)^n \approx \frac{1}{en!} \quad (26)$$

for $N \gg n$. The detection of more than one excited atom would violate energy conservation.

We are led to the important conclusion that inconsistencies arise when considering a classical pulse incident on a quantum mechanical medium when the energy in the pulse is on the order of the excitation energy of a single atom in the medium. These inconsistencies point to the need for a quantized field description. The advantage of treating the absorption of a classical pulse over the photoelectric effect to show that field quantization is needed is that probe absorption represents a fully solvable model in which we can see directly the way in which a classical field assumption leads to physically inconsistent results.

III. QUANTIZED COHERENT STATE PULSE ABSORPTION

Many of the features of the classical pulse problem are reproduced if the classical field is replaced by a multimode quantum coherent state of the field having average energy $\hbar\omega_0$. It is not necessary to go into a detailed calculation of the effects of such a pulse on the medium, although such a calculation can be done following the method used in Ref. 6. Instead, we need only to look at the properties of the incident field at $t=0$ to understand the dynamics of the problem.

We consider an effective one-dimensional problem corresponding to a pulse having cross-sectional area A propagating in the z direction with polarization $\hat{\mathbf{x}}$. The positive frequency component of the quantized electric field is

$$\mathbf{E}^+(Z) = i\hat{\mathbf{x}} \sum_j \left(\frac{\hbar\omega_j}{2\epsilon_0 AL}\right)^{1/2} a_j e^{ik_j Z}, \quad (27)$$

where $\omega_j = k_j c$ is the frequency of mode j , a_j is the destruction operator for mode j , and AL is the quantization volume. With periodic boundary conditions, $k_j = 2\pi j/L$, where j is an integer (positive, negative, or zero). The energy in the field is

$$2A\epsilon_0 \int_{-L/2}^{L/2} \mathbf{E}_j^-(Z) \cdot \mathbf{E}_j^+(Z) dZ = \hbar\omega_j a_j^\dagger a_j \delta_{jj'}. \quad (28)$$

That is, the expectation value of the energy in mode j of the field is equal to $n_j \hbar\omega_j$, where n_j is the average number of photons in this mode. For future reference, we note that the prescription for transforming from discrete to continuum modes of the field is

$$\sum_j \rightarrow \frac{L}{2\pi} \int_{-\infty}^{\infty} dk. \quad (29)$$

The initial state of the field is taken to be the multimode coherent state, which is denoted by

$$|\psi\rangle = |\alpha_1, \alpha_2, \alpha_3, \dots\rangle = \prod_j |\alpha_j\rangle, \quad (30)$$

where

$$|\alpha_j\rangle = \sum_{n_j=0}^{\infty} \frac{(\alpha_j)^{n_j} e^{-|\alpha_j|^2/2}}{\sqrt{n_j!}} |n_j\rangle. \quad (31)$$

Here, α_j are chosen so that the expectation value of the electric field operator at $t=0$ coincides with the initial classical field given in Eq. (5).

It follows from Eqs. (6), (27), and (29) that the expectation value of the field is

$$\langle \mathbf{E}^+(Z, t=0) \rangle = i\hat{\mathbf{x}} \sum_j \prod_{\ell, \ell'} \left(\frac{\hbar\omega_j}{2\epsilon_0 AL}\right)^{1/2} e^{ik_j Z} \langle \alpha_\ell | a_j | \alpha_{\ell'} \rangle \quad (32)$$

$$= i\hat{\mathbf{x}} \sum_j \left(\frac{\hbar\omega_j}{2\epsilon_0 AL}\right)^{1/2} \alpha_j e^{ik_j Z} \quad (33)$$

$$\rightarrow i\hat{\mathbf{x}} \frac{L}{2\pi} \int_{-\infty}^{\infty} dk \left(\frac{\hbar kc}{2\epsilon_0 AL}\right)^{1/2} \alpha_k e^{ikZ}. \quad (34)$$

In contrast, from Eqs. (5) and (6), we find that the classical pulse amplitude is

$$\begin{aligned} \mathbf{E}^+(Z, t=0) &= \frac{1}{2} E_0 \hat{\mathbf{x}} e^{ik_0 Z} f(Z+Z_0) \\ &= \frac{1}{2} E_0 \hat{\mathbf{x}} e^{ik_0 Z} e^{-(Z+Z_0)^2/2(\delta Z)^2} \end{aligned} \quad (35)$$

$$\begin{aligned} &= \frac{1}{2} \hat{\mathbf{x}} \left(\frac{2\hbar\omega_0}{\epsilon_0 A (\delta Z) \sqrt{\pi}}\right)^{1/2} \left(\frac{\delta Z}{\sqrt{2\pi}}\right) e^{-ik_0 Z_0} \\ &\quad \times \int_{-\infty}^{\infty} dk e^{-(k-k_0)^2(\delta Z)^2/2} e^{ik(Z+Z_0)}, \end{aligned} \quad (36)$$

where E_0 was taken from Eq. (8). If we compare Eqs. (34) and (36), we obtain

$$\alpha_k = -i \left(\frac{2\omega_0(\delta Z)\sqrt{\pi}}{kcL}\right)^{1/2} e^{-(k-k_0)^2(\delta Z)^2/2} e^{i(k-k_0)Z_0}. \quad (37)$$

With this value of α_k , the average energy in the field of this multimode state is equal to

$$\langle W \rangle = \sum_j \hbar\omega_j |\alpha_j|^2 \rightarrow \frac{Lc}{2\pi} \int_{-\infty}^{\infty} \hbar k |\alpha_k|^2 dk = \hbar\omega_0, \quad (38)$$

as desired.

When this field enters the sample, it is totally absorbed, just as in the classical case. As a consequence, irrespective of the atom-field dynamics, the probability that n atoms are excited is equal to the probability $P(n)$ that there are n photons in the incident field pulse. There is a finite probability $P(0)$ that the quantized multimode coherent state field has no photons. If we expand the initial state of the field in the number state basis as

$$|\psi\rangle = |\alpha_1, \alpha_2, \alpha_3, \dots\rangle = \prod_j \sum_{n_j=0}^{\infty} \frac{(\alpha_j)^{n_j} e^{-|\alpha_j|^2/2}}{\sqrt{n_j!}} |n_j\rangle, \quad (39)$$

it is not difficult to calculate

$$P(0) = \exp\left(-\sum |\alpha_j|^2\right) \rightarrow \exp\left(-\frac{L}{2\pi} \int_{-\infty}^{\infty} |\alpha_k|^2 dk\right) = \frac{1}{e}, \quad (40)$$

where Eq. (37) was used with k evaluated at k_0 .

There is a probability equal to $1/e$ that the quantized multimode coherent state contains no photons. In other words, if we repeat the experiment of sending this field into the sample many times, no field is sent into the sample a fraction $1/e$ of the times.

We can now understand the relation of this result to the classical pulse case. For both classical and quantized fields, the probability that no atoms are excited is identical. However, in the classical case, this result leads to a contradiction because the incoming pulse has a well-defined energy equal to $\hbar\omega_0$. In the quantized field case, there is no contradiction because the field is not in an eigenstate of the energy for the field. The average field energy is equal to $\hbar\omega_0$, but there is a $1/e$ probability of having no photons in the field, which maps into the corresponding probability to have no atoms excited.

Similarly, we can show that

$$P(n) = \frac{1}{n!e} \sum_{m,m',m'',\dots=1}^N |\alpha_m|^2 |\alpha_{m'}|^2 |\alpha_{m''}|^2 \cdots \epsilon_{m,m',m'',\dots} \quad (41)$$

$$\approx \frac{1}{n!e} \left(\frac{L}{2\pi} \int_{-\infty}^{\infty} |\alpha_k|^2 dk \right)^n = \frac{1}{n!e} \quad (42)$$

where the probability to excite two photons in the same mode is taken to be negligibly small. Equation (42) is consistent with the classical field result for the excitation of n atoms in the sample. Again, there is no inconsistency with energy conservation for the quantized field case because the field is not in an energy eigenstate, and there is some probability that the initial field pulse contains more than $\hbar\omega_0$ of energy. On average, $\hbar\omega_0$ is transferred to the atoms. However, sometimes no energy is transferred and other times $n\hbar\omega_0$ ($n \geq 1$) is transferred.

IV. CONCLUSION

We have shown that by considering the seemingly innocuous problem of optical pulse absorption by an optically dense medium, we are led to the conclusion that the radiation field must be quantized if we are to avoid results that are inconsistent with reality. Although we made a highly restricted set

of assumptions for our calculation, we did so only to simplify the calculations and to illustrate the relevant physics. It is not difficult to argue that inconsistencies arise whenever any classical field pulse leaves atoms or a quantized material system in a superposition of energy eigenstates. Because the atoms are not left in an eigenstate of energy and because the classical field pulse always has a definite energy, the entire atom-field system no longer conserves energy. For example, if a classical pulse interacts with a single two-level atom and leaves it in an equal superposition of its ground and excited states (separated by frequency ω_0), the classical field energy must be reduced by $\hbar\omega_0/2$. However, if we measure the atom in its ground state following the interaction, the energy in the field pulse should be unchanged and not reduced at all. We are led naturally to a contradiction because the final state for our system cannot be an entangled state of the atom and the (classical) field. The use of a quantized field state allows for such entanglement.

In the article by Lamb and Scully,² as in Einstein's original paper,³ it was assumed that any change in the energy in the field produced by the medium could be ignored—in this limit, a classical field description is valid. It would be interesting to see if an analogous argument for field quantization could be made without reference to a quantized material system.

ACKNOWLEDGMENTS

One of the authors (P.R.B.) would like to thank A. Leanhardt, H. Deng, G. W. Ford, and A. Rojo for the helpful discussions.

¹See, for example, the discussion given in J. C. Garrison and R. Y. Chiao, *Quantum Optics* (Oxford U. P., Oxford, 2008), Chap. 1.

²W. E. Lamb, Jr. and M. O. Scully, "The photoelectric effect without photons," *Polarisation, Matière et Rayonnement* (Presses U. de France, Paris, 1969), pp. 363–369.

³A. Einstein, "Concerning an heuristic point of view toward the emission and transformation of light," *Ann. Phys.* **332**, 132–148 (1905); an English translation of this article is given in A. B. Arons and M. B. Peppard, "Einstein's Proposal of the Photon Concept—a Translation of the Annalen der Physik Paper of 1905," *Am. J. Phys.* **33**, 367–374 (1965).

⁴Equation (4) can be replaced by the weaker condition, $\rho\lambda_0^3\gamma/\Delta \ll 1$, for our inhomogeneously broadened sample.

⁵C. W. Thiel, Y. Sun, T. Böttger, W. R. Babbitt, and R. L. Cone, "Optical decoherence and persistent spectral hole burning in $\text{Tm}^{3+}:\text{LiNbO}_3$," *J. Lumin.* **130**, 1598–1602 (2010). The inhomogeneous width is on the order of 300 GHz and the transverse relaxation rate is on the order of 1.4 μs for a 794 nm transition in the Tm^{3+} ion.

⁶P. R. Berman and J.-L. Le Gouët, "Quantum information storage: A Schrödinger picture approach," *Phys. Rev. A* **79**, 042314 (2009).