On Pancharatnam's quantum theory of dispersion

D. T. Pegg

Faculty of Science and Technology, Griffith University, Nathan, Brisbane, Q 4111, Australia

Pancharatnam considered a quantum mechanical system consisting of *N* atoms interacting with an off-resonance one-photon field. He showed that the light shift is simply related to the atomic polarizability and gave a quantum mechanical interpretation of the refractive index. Here we extend his work to include fields in arbitrary superpositions of photon number states and describe the phenomenon in terms of the concept of a dressed *field*, which complements the dressed atom concept.

MY association with Pancharatnam began when I arrived at the Clarendon Laboratory, Oxford, in the beginning of 1968 and the aspects of his work with which I became most familiar were those subsequently published posthumously by G. W. Series. The paper which has had a special appeal to me is the quantum theory of dispersion in relation to light shifts[1]. One reason for this is that Panch gave me a copy of his notes of this paper for comment and, after I felt I understood the work sufficiently, I made a note of some points which I intended to discuss with him. Unfortunately this discussion was precluded by his illness and untimely death, after which I gave his notes to Professor Series who prepared them for publication. As mentioned by Professor Series in the final published version, the work was incomplete and I have wondered how Panch would have completed or extended it. Another reason for the special appeal concerns Panch's concept of a state representing a 'photon in the polarized medium'. About that time I was becoming aware that my own work involving semiclassical physics was related to the fully quantum mechanical dressed-atom approach used by Cohen-Tannoudji and Haroche[2,3], which has gained an important and useful place in quantum optics. It occurred to me that the photon in the polarized medium might also be able to be described in terms of a dressed field, that is, a field dressed by the atoms of the medium. If so, this would help me see more clearly the relationship he established between dispersion and the light shift. Without speculating whether Panch would have continued in this direction, I explore in this paper the dressed-field description while extending his work to include more than the one-photon state, allowing fields which are in arbitrary superpositions of photon number states to be considered.

Pancharatnam's approach

The system considered by Pancharatnam[1] was that of radiation in a unit volume interacting with N independent atoms which constitute the medium. The state space of the system is spanned by the infinite number of unperturbed energy eigenstates, that is eigenstates of the Hamiltonian in which the interaction is set to zero, but only a subspace of states is considered. Specifically, these include the unperturbed eigenstates $|\boldsymbol{K}_e, G\rangle$ and $|0, S_m\rangle$. The first describes one photon with a particular wave-vector and polarization $\boldsymbol{e}$, with no photons in any other mode, and with all atoms in their electronic ground states. The second describes no photons in any mode and the Sth atom in an excited state m. This limited approach does not include spontaneous emission, but Pancharatnam allows for this by inserting an imaginary part $\mathrm{i}\,\Gamma\hbar/2$ into the excited state energy to describe radiative damping. The decay of the upper state was an area which he indicated he would explore further.

The fundamental time-dependent process of interest is the transition $|\boldsymbol{K}_e, G\rangle \to |0, S_m\rangle \to |\boldsymbol{K}_e, G\rangle$, representing a virtual absorption of the photon $\hbar\omega$, with one atom S raised to an excited state m followed by the re-emission, or forward scattering, of the photon. The language used in the paper, however, is that of time-independent perturbation theory. Just as the eigenstate $|\boldsymbol{K}_e, G\rangle$ of the Hamiltonian without the interaction term represents a photon in the unpolarized medium, the corresponding perturbed state $|\boldsymbol{K}_e, G\rangle_{\rm med}$, which approximately diagonalizes the Hamiltonian of the subsystem including the interaction, represents a 'photon in the polarized medium'.

Pancharatnam finds an approximate steady-state solution of the Schrödinger equation of the form $|\boldsymbol{K}_e, G\rangle_{\rm med}\exp[-\mathrm{i}(\bar{E}t/\hbar)]$ which reduces in the absence of interaction to $|\boldsymbol{K}_e, G\rangle\exp(-\mathrm{i}\,\omega t)$, the solution of the unperturbed Schrödinger equation. The level shift of $|\boldsymbol{K}_e, G\rangle_{\rm med}$ away from $|\boldsymbol{K}_e, G\rangle$ is $\hbar\varpi = \bar{E} - \hbar\omega$, which is found to be $-2\pi\alpha_{ee}\,\hbar\omega$ where

$$\alpha_{ee} = \sum_{S,m}\hbar^{-1}|\langle m\,|\hat{P}_S\cdot\boldsymbol{e}\,|g\rangle|^2/[\omega_m(S)-\omega-\mathrm{i}\,\Gamma/2]. \quad (1)$$

This shows the direct connection between the light shift and the polarizability. Here $\hat{\boldsymbol{P}}_S$ and $\hbar\omega_m(S)$ are the dipole-moment operator and the energy of state m for the atom S. An equation equivalent to this was previously obtained by Pancharatnam[4] by a semiclassical approach. The expression for the light shift is equivalent to the expression first derived by Barrat and Cohen-Tannoudji[5].

To relate the level shift to dispersion, we might assume that the velocity of a photon, which in the absence of interaction with the medium is simply ω/K where $\hbar K$ is the momentum, simply becomes $\bar{v} = \bar{\omega}'/K$ where $\hbar\bar{\omega}'$ is the energy difference between the state $|K_e, G\rangle_{med}$ with one photon and $|0, G\rangle$ with none. This would give for the refractive index $\bar{n}$:

$$(\bar{n}-1) = \frac{c-\bar{v}}{\bar{v}} = \frac{\omega-\bar{\omega}'}{\bar{\omega}'} \approx -\frac{\delta\bar{\omega}}{\omega}, \tag{2}$$

which for small $(\bar{n}-1)$ is the same as that derived by semiclassical theory. Pancharatnam, however, points out that it is not clear why we should be able to do this. He therefore also presents as argument based on the expectation value of the Heisenberg first-order correlation operator and shows that this expectation value is the same as that for a classical field which varies as $\exp[\mathrm{i}(K \cdot r - \bar{\omega}' t)]$. Thus $\bar{\omega}'/K$ plays the role of the phase velocity.

General fields

Pancharatnam's limitation of the state space restricts the fields which can be studied to superpositions of the one and zero photon states. Here we generalize to include more general fields, that is, fields in an arbitrary superposition of photon number states. We shall not be concerned with the detailed expression for the polarizability in terms of the dipole-moment operators, nor with the specification of the polarization of the field. Instead, we shall simplify the problem, firstly by considering just a single two-level atom at position r with ground and excited states $|g\rangle$ and $|e\rangle$ interacting with a field in a single mode, as expressed by the Hamiltonian

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_a + \hat{V} + \mathcal{H}_f \\ &= |e\rangle\langle e|\hbar\omega_0 + \mathrm{i}(\hat{a}^\dagger \exp(-\mathrm{i}k \cdot r)|g\rangle\langle e| \\ &\quad - h.c.)\hbar\Omega + \hat{N}\hbar\omega,\end{aligned} \tag{3}$$

where $\mathcal{H}_a$ and $\mathcal{H}_f$ represent the free atom and free field Hamiltonians, $\hat{N}$ is the photon number operator with eigenstates $|n\rangle$ and the energies of $|g\rangle$ and $|e\rangle$ are zero and $\hbar\omega_0$. We have written the interaction term in the rotating wave approximation, where $\hat{a}$ and $\hat{a}^\dagger$ are the photon annihilation and creation operators. The strength of the coupling is given by $\hbar\Omega$ and is dependent on the dipole of the atom. Of course, by ignoring the other modes, we are also not including spontaneous emission. We shall simply disregard this for our present discussion in which we are effectively assuming that the medium is transparent.

The electric field in the Schrödinger (time-independent) picture we are using is represented by the operator[1] $\hat{E}(r)$ which is proportional to $\mathrm{i}[\hat{a}\exp(\mathrm{i}k \cdot r) - h.c.]$. The propagation is best viewed in the Heisenberg picture, in which the field operator is, in the absence of coupling, that is for $\hat{V} = 0$,

$$\begin{aligned}\hat{E}(r, t) &= \exp(\mathrm{i}\mathcal{H}t/\hbar)\,\hat{E}(r)\,(\exp(-\mathrm{i}\mathcal{H}t/\hbar)), \\ &\propto \mathrm{i}[\hat{a}(t)\exp(-\mathrm{i}\,\omega t + \mathrm{i}k \cdot r) - h.c.].\end{aligned} \tag{4}$$

This represents a wave with phase velocity $c = \omega/k$. Also when $\hat{V} = 0$, the eigenstates of $\mathcal{H}$ are $|n\rangle|g\rangle$ and $|n\rangle|e\rangle$. The former have energies $n\hbar\omega$ and the latter have energies of $\hbar(\omega_0 + n\omega)$.

We now assume that the light is sufficiently off resonance to allow us to diagonalize $\mathcal{H}$ by perturbation theory, but not so far off resonance as to invalidate the rotating wave approximation. The new energy eigenvalues are obtainable from the eigenvalues E_p^0 for the unperturbed states $|p\rangle$ by use of the second order perturbation shift, or repulsion, formula[6]

$$E_p = E_p^0 - \sum_{k \neq p} |\langle k|\hat{V}|p\rangle|^2 (E_k^0 - E_p^0)^{-1}. \tag{5}$$

The new eigenstates are related to the unperturbed states by[6]

$$|E_p\rangle = |p\rangle - \sum_{k \neq p} \langle k|\hat{V}|p\rangle (E_k^0 - E_p^0)^{-1} |k\rangle. \tag{6}$$

For the atom plus field system, we shall denote by E_{ng} and E_{ne} the energies of the perturbed states $|n, g\rangle_{pert}$ and $|n, e\rangle_{pert}$ which correspond to the unperturbed states $|n\rangle|g\rangle$ and $|n\rangle|e\rangle$.

From equation (3) $\hat{V}$ couples $|n\rangle|g\rangle$ and $|n-1\rangle|e\rangle$ with a matrix element of modulus $n^{1/2}\hbar|\Omega|$, giving

$$E_{ng} = n\hbar[\omega - \Omega^2/(\omega_0 - \omega)] \tag{7}$$

and

$$\begin{aligned}E_{ne} &= \hbar[\omega_0 + \Omega^2/(\omega_0 - \omega)] \\ &\quad + n\hbar[\omega + \Omega^2/(\omega_0 - \omega)].\end{aligned} \tag{8}$$

For our present discussion, it is sufficient to use equation (6) to write the perturbed states $|n, g\rangle_{pert}$ as $|n\rangle|g\rangle + O(\Omega)$ where the second term is of order Ω and contains the state $|n-1\rangle|e\rangle$. A similar expression holds for $|n, e\rangle_{pert}$.

So far we have not specified the initial state of the atom or of the field. We now digress a little. Suppose we specify the initial state of the field. For example, let it be in a coherent state of reasonably high intensity. Such a state has a spread of photon number much smaller than the mean $\langle n \rangle$. By specifying the state of the field we can reduce the states of the total system to those of relevance, which in this case will be the states involving photon numbers n clustered around $\langle n \rangle$. Consequently, the energies of states $|n, e\rangle_{\text{pert}}$ and $|n, g\rangle_{\text{pert}}$ which are of interest differ from their unperturbed values by amounts of approximately $+\Delta$ and $-\Delta$, respectively, where $\Delta = \langle n \rangle \hbar\Omega^2/(\omega_0 - \omega)$. This means that the relevant energy eigenvalues of the atom plus field system can be written to a good approximation as $\hbar\omega_0 + \Delta + n\hbar\omega$ and $-\Delta + n\hbar\omega$. These are the same as those of a two level atom with excited and ground state energies of $\hbar\omega_0 + \Delta$ and $-\Delta$, together with, but not interacting with, a field with energy eigenvalues $n\hbar\omega$. Thus if we are interested in making measurements on the atom, for example by probing it with a second field, we can account for the interaction with the first field by treating the atom plus field system as a 'dressed atom' with these new energy levels[3]. This relates Δ simply to the light shift.

The case of interest to us is just the opposite to that above. We wish to specify the initial state of the atom and not of the field. We also wish to examine possible measurements on the field and not on the atom. Thus it is more appropriate to speak of the atom plus field system as a 'dressed field' that is, a field dressed by an atom in state $|g\rangle$. This specification of the state of the atom reduces the states of the total system to those of relevance, which in this case will be $|n, g\rangle_{\text{pert}}$. The relevant energy levels of the system are thus E_{ng} in equation (7), that is $n\hbar\omega_g$ where

$$\omega_g = \omega - \Omega^2/(\omega_0 - \omega). \tag{9}$$

These are the same energy levels as for a field of frequency ω_g together with, but not interacting with, an atom in state $|g\rangle$. Thus when we examine properties of the field we can account for the interaction with the atom by using dressed energy states, or a dressed frequency, of the field. The perturbation shifts of the total atom plus field system could be referred to here as 'atom' shifts, that is, shifts in the field energy states caused by the interaction with the atom, whereas in the dressed atom case they would be referred to as light shifts of the atom. The important point is that they are the same shifts.

Our next step is to generalize to a field dressed by a medium comprising N two-level atoms in a unit volume. We write the excited state energy and the coupling for the Sth atom as $\hbar\omega_S$ and $\hbar\Omega_S$ and call them the ground state of the medium, with all the atoms in the zero energy state, $|G\rangle$. We are interested in the field dressed by the medium in this state. Omitting the details, we find that the energy of the perturbed state $|n, G\rangle_{\text{med}}$, which corresponds to the unperturbed state $|n\rangle\,|G\rangle$, is $n\hbar\omega_G$ where

$$\omega_G = \omega - \sum_S \Omega_S^2/(\omega_S - \omega). \tag{10}$$

This is the same as the energy of an n-photon field of frequency ω_G not interacting with the medium. We might therefore associate the frequency of the dressed field with ω_G, and, following Pancharatnam[1], simply write for the refractive index $\bar{n}$:

$$\bar{n} - 1 = \frac{\omega - \omega_G}{\omega_G}. \tag{11}$$

Again, however, we need some argument to confirm this.

At time zero, let the medium be in the ground state $|G\rangle$ and the field in some state $|f_0\rangle$. We wish to examine the state of the field after it has interacted with the medium for a time t. In general, the state of the medium plus field system at time t will not factorize into a state of the field and a state of the medium. Instead it will be an entangled, or correlated, state consisting of the sum of products of states of the field and of the medium. We can assume, however, that the interaction is sufficiently small and off resonance for us to retain only the dominant term in the entanglement, which will be of the form $|G\rangle\,|f(t)\rangle$. This is equivalent to specifying that the fundamental process is that which leaves the medium in its ground state, that is, that the transition involved is a virtual absorption of a photon followed by re-emission, or forward scattering of the photon. The net absorption of light by the medium is zero. Thus we are interested in calculating $|f(t)\rangle$ where the general state of the medium plus field at time t is given by

$$U(t, 0)|G\rangle|f_0\rangle = |G\rangle|f(t)\rangle + \text{other terms}, \tag{12}$$

with $U(t, 0)$ being the time displacement operator. We let $\mathcal{H}$ now be the Hamiltonian for the system comprising the N atoms interacting with the field. Substituting $\exp(-\mathrm{i}\,\mathcal{H}t/\hbar)$ for $U(t, 0)$, and projecting both sides of equation (12) onto state $\langle G|$ gives

$$\begin{aligned} |f(t)\rangle &= \langle G|\exp(-\mathrm{i}\,\mathcal{H}t/\hbar)|G\rangle|f_0\rangle \\ &= U_{\text{med}}(t, 0)|f_0\rangle \end{aligned} \tag{13}$$

say. In terms of the complete set of eigenstates $|E_p\rangle$ of $\mathcal{H}$, we have

$$U_{\text{med}}(t, 0) = \sum_p \langle G|\exp(-\mathrm{i}\mathcal{H}t/\hbar)\,|E_p\rangle\langle E_p|G\rangle. \tag{14}$$

From equation (6), $|G, n\rangle_{\text{med}}$ is $|G\rangle\,|n\rangle + O(\Omega)$ where the second term, of order Ω, contains states orthogonal to $|G\rangle$. Thus

$$\langle G \mid G, n\rangle_{\text{med}} = \mid n\rangle. \tag{15}$$

States $|E_p\rangle$ other than $|G, n\rangle_{\text{med}}$ contain a component of $|G\rangle|n\rangle$, but only to order Ω, so $\langle G|E_p\rangle$ for these states will be of order Ω. Thus equation (14) will contain a sum of terms of which the dominant one is for $|E_p\rangle = |G, n\rangle_{\text{med}}$, and which is given by

$$\sum_n \langle G|\exp(-\mathrm{i}\mathcal{H}t/\hbar)\,|G,n\rangle_{\text{med}}\,\langle n|$$

$$= \sum_n \langle G|G, n\rangle_{\text{med}}\,\langle n|\exp(-\mathrm{i}n\omega_G\, t)$$

$$= \sum_n |n\rangle\,\langle n|\exp(-\mathrm{i}n\omega_G\, t). \tag{16}$$

Terms involving states $|E_p\rangle$ other than $|G, n\rangle_{\text{med}}$ will contain at most a component of $|G, n\rangle$ of order Ω, and will lead to terms in U_{med} only of order Ω^2 at most. Including these gives

$$U_{\text{med}}(t, 0) = \exp(-\mathrm{i}\,\hat{N}\,\omega_G\, t) + O(\Omega^2). \tag{17}$$

The equality of equation (16) and the first term of equation (17) is easy to check by expanding the series and using $\sum n\,|n\rangle\langle n|$ for $\hat{N}$. Comparison of the dominant term of equation (17) with the time evolution operator of the free field $\exp(-\mathrm{i}\,\mathcal{H}_f t/\hbar)$ for which $\mathcal{H}_f = \hat{N}\hbar\omega$ shows that the field evolves approximately in the medium as if it were a free field of frequency ω_G. Specifically, the Heisenberg field operator $U_{\text{med}}^{-1}\hat{E}(\boldsymbol{r})U_{\text{med}}$ becomes proportional to $\mathrm{i}(\hat{a}\exp(-\mathrm{i}\,\omega_G\, t + \mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{r}) - h.c.)$, which is the expression for a wave with a phase velocity of ω_G/k. Thus equation (11) is confirmed. The terms of order Ω^2 in equation (17) will add substantially smaller components of the field with different frequencies.

Conclusion

It is possible to remove the restriction of the rotating wave approximation in equation (3) and obtain a result closer to the classical case, but this will not be done here. From what has been done it is clear that Pancharatnam's quantum theory of dispersion[1] for a one-photon field can be extended to include fields in arbitrary superpositions of photon number states. In situations where the initial state of the field is specified and the evolution of the atom is examined, the total system of an atom plus interacting field can be considered as a dressed atom. Our case, however, is the opposite. The initial state of the medium, that is of the atoms, is specified and we are interested in the evolution of the field. Pancharatnam's concept of a 'photon in the polarized medium' corresponds to considering the total system of atoms interacting with the field as a dressed field. The dominant component of the dressed field evolves in the medium as though it were a free field with frequency ω_G. The associated photon energy shift $\hbar(\omega - \omega_G)$ is obtainable directly from the energy eigenstates of the atoms plus field system. The underlying relation of dispersion to the light shifts arises because the shifts of the perturbed energy levels of the complete system from their unperturbed values are the same whether the system is interpreted as dressed atoms exhibiting light shifts, or as a dressed field exhibiting atom shifts.

Although the basic concepts presented here are inherent in Panch's paper, I do not know whether or not he would have extended his work in this particular direction. However, I like to think he may have.

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