

IS THERE A PROBLEM WITH OUR HAMILTONIANS FOR QUANTUM NONLINEAR OPTICAL PROCESSES?

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Abstract

The models we use, habitually, to describe quantum nonlinear optical processes have been remarkably successful, yet, with few exceptions, they each contain a mathematical flaw. We present this flaw, show how it can be fixed, and, in the process, suggest why we can continue to use our favored Hamiltonians.

Keywords: quantum optics, nonlinear optics, model Hamiltonians, three-wave mixing.

1. Introduction: a Dilemma

Quantum nonlinear optics is now very well developed, with photonic devices such as parametric oscillators and spontaneous parametric down-converters playing the role of workhorses in experimental demonstrations of exotic quantum phenomena (including entanglement) and in the advance of quantum information technology. Of particular relevance to us are those associated with second- and third-order nonlinearities corresponding to the interaction between three or four fields. We shall see that the natural and widely employed quantum descriptions of these processes rely on Hamiltonians with spectra that are unbounded from below [1]. There are very good mathematical and, indeed physical, reasons for doubting the validity of such an unbounded Hamiltonian and these doubts, in turn, challenge our confidence in our understanding of quantum nonlinear optical processes. The problem is an old one, although perhaps not well known, and it is for this reason, primarily, that this article cites mostly books rather than original papers; tracking down the full set of relevant papers published over the last 30 or so years would be an exhausting challenge.

Let us begin by presenting the problem as simply as possible. To this end, we consider a simple model of an intracavity optical parametric oscillator, in which a nonlinear crystal mediates the reversible transformation of single photons from mode a into a pair of photons, one each in modes b and c . A simple Hamiltonian used to describe this process might be of the form

$$\hat{H} = \omega_a \hat{a}^\dagger \hat{a} + \omega_b \hat{b}^\dagger \hat{b} + \omega_c \hat{c}^\dagger \hat{c} + \kappa \left(\hat{a}^\dagger \hat{b} \hat{c} + \hat{b}^\dagger \hat{c}^\dagger \hat{a} \right), \quad (1)$$

where, as is common practice in quantum optics, we have chosen units such that $\hbar = 1$. We shall assume that the parametric process is resonant so that $\omega_a = \omega_b + \omega_c$. This is not strictly necessary but making this choice simplifies the analysis without affecting the points we seek to make. There is a very large variety of models of this form; these include four-wave processes in which there is an additional mode, with annihilation operator \hat{d} , and for which \hat{a} is replaced by $\hat{a}\hat{d}$ in the interaction term, models in which

one or more modes are strong and treated classically, and also models using continuum modes. These are often supplemented by driving terms and the ubiquitous losses and noise. The treatment of these is now the domain of specialist textbooks devoted to quantum optics [2–11]. That our simple Hamiltonian has a spectrum that is unbounded from below is most readily demonstrated by evaluating the expectation value of \hat{H} for the three-mode coherent state $|\alpha\rangle_a|\beta\rangle_b|\gamma\rangle_c$ [1],

$$\langle\hat{H}\rangle = \omega_a|\alpha|^2 + \omega_b|\beta|^2 + \omega_c|\gamma|^2 + \kappa(\alpha^*\beta\gamma + \beta^*\gamma^*\alpha), \quad (2)$$

where we used the familiar eigenstate property of the coherent states: $\hat{a}|\alpha\rangle = \alpha|\alpha\rangle$ [8]. This expectation value can take any value and, in particular, any negative value as may readily be seen by setting $|\alpha| = |\beta| = |\gamma|$ and choosing the phases of these such that the interaction term is negative,

$$\langle\hat{H}\rangle = (\omega_a + \omega_b + \omega_c)|\alpha|^2 - 2|\kappa||\alpha|^3, \quad (3)$$

which tends to $-\infty$ for large $|\alpha|$. The expectation value of the Hamiltonian cannot be less than its lowest energy eigenvalue, and it follows, therefore, that the Hamiltonian has a spectrum that is unbounded from below.

There are very good reasons for being suspicious of and even rejecting Hamiltonians with no ground state. Perhaps the most telling of these is the possibility, were such a system to be realized, of extracting unbounded amounts of energy associated with the decay of the system to every lower energy states.

2. Resolution I: Restricted State Space

The first thing to notice about the above argument is that the runaway behavior towards negative energy eigenstates sets in at very high photon numbers corresponding to high optical, electric field strengths. As an indication of this, we can write our polarization as a nonlinear function of the electric field [12, 13],

$$P = \varepsilon_0 \left(\chi^{(1)}E + \chi^{(2)}E^2 + \dots \right). \quad (4)$$

Typical values of the nonlinear susceptibility, $\chi^{(2)}$, are in the range 10^{-11} to 10^{-12} $\text{m} \cdot \text{V}^{-1}$ [12], and this suggests that we need an optical electric field strength in the region of perhaps 10^{12} $\text{V} \cdot \text{m}^{-1}$ corresponding to an intensity of the order of 10^{18} $\text{W} \cdot \text{cm}^{-2}$ before the nonlinear susceptibility dominates and perhaps leads to the problems indicated. So one might very well take the view that the problem does not arise in the experimental regime of interest. This is not quite satisfactory, however, unless we can show that the dynamical evolution of the modes cannot take us into the regime in which the unbounded negative-energy eigenspace occurs.

There is, thankfully, a natural set of conservation relations that apply, and these ensure that if our initial state has no overlap with the troublesome negative energy eigenstates, then it will not acquire one. It may be shown, either by inspection of the Hamiltonian or by explicit calculation, that there are three conserved quantities corresponding to the operators

$$\hat{M}_1 = \hat{b}^\dagger\hat{b} + \hat{a}^\dagger\hat{a}, \quad \hat{M}_2 = \hat{c}^\dagger\hat{c} + \hat{a}^\dagger\hat{a}, \quad (5)$$

with the third following from these:

$$\hat{M}_3 = \hat{c}^\dagger\hat{c} - \hat{b}^\dagger\hat{b}. \quad (6)$$

These three are the operator analogs of the Manley–Rowe relations familiar from classical nonlinear optics [12,13]. It follows that we can divide the state space into noninteracting blocks, each characterized by a pair of positive integers, M_1 and M_2 , corresponding to the first of our two conserved quantities. It then follows that within each block, the number of photons in any mode can never exceed the largest of these two integers. This procedure provides a very natural way to solve for the dynamics of the three-mode state and of its mathematically equivalent model of a number of two level-atoms interacting cooperatively with a single cavity mode [14–16].

In light of the above observations, it is interesting, at least mathematically, to ask how it is that a Hamiltonian with photon-number conservation laws can lead to an energy eigenvalue spectrum that is unbounded from below, as surely adding more photons will increase the energy. In fact, this is not so, as may readily be seen in the following estimate. Let us suppose that we have a low-energy eigenstate, in which all three modes have roughly N photons but with some small variations in the superposition of photon-number product states. For this state, we can estimate the energy eigenvalue by replacing each of the creation and annihilation operators by \sqrt{N} and suitably selecting the phases in the superposition such that the contribution from the interaction term is negative. We find

$$E_N \approx 2N\omega_a - 2|\kappa|N^{3/2}, \quad (7)$$

so that the energy becomes negative for $\sqrt{N} > \omega_a/|\kappa|$. It will become increasingly negative as we *increase* N so that adding photons in this regime will *reduce* the overall energy for this state. If we restrict our state space to contain only photon numbers very much less than this value, then using our coupled-mode Hamiltonian will not get us into the difficulties associated with much higher photon numbers. This means that we will avoid problems if we restrict our Hamiltonian as being valid *only* for photon numbers below some upper limit and restrict our state space to be spanned only by photon numbers less than this upper value.

3. Resolution II: Higher-Order Processes

The resolution presented in the preceding section is somewhat mathematical in nature and, as a counterbalance, we present here a more physical line of reasoning. We have seen that the problem of unbounded negative energy eigenvalues arises at high field strengths, corresponding to very high photon numbers, and this suggests that higher-order nonlinear optical processes will come into play before the problem is reached. If so, then the physical resolution will be a more accurate Hamiltonian that does not have the problem of unbounded negative-energy eigenstates. This does indeed turn out to be the case.

Let us consider a three-level atomic model with a ground state $|0\rangle$ and two excited states, $|1\rangle$ and $|2\rangle$, coupled by our three optical modes, as depicted in Fig. 1.* Such level schemes form the basis of microscopic calculations of nonlinear susceptibilities and can be found in many texts on nonlinear optics [12,13,17–21]. It suffices, for our purposes, to consider just a single atom as our nonlinear medium; including many atoms to form a nonlinear medium presents no special difficulties but would add an unnecessary complication. Our Hamiltonian has the form

$$\hat{H} = \hat{H}_F + \hat{H}_A + \hat{V}, \quad (8)$$

*Strictly speaking, we need an additional electric field to mix the parities of the energy levels so that the pattern of transitions are all allowed.

where the component parts are

$$\begin{aligned}\hat{H}_F &= \omega_a \hat{a}^\dagger \hat{a} + \omega_b \hat{b}^\dagger \hat{b} + \omega_c \hat{c}^\dagger \hat{c}, & \hat{H}_A &= \sum_{i=0}^2 \mathcal{E}_i |i\rangle \langle i|, \\ \hat{V} &= g_a \left(|2\rangle \langle 0| \hat{a} + \hat{a}^\dagger |0\rangle \langle 2| \right) + g_c \left(|2\rangle \langle 1| \hat{c} + \hat{c}^\dagger |1\rangle \langle 2| \right) + g_b \left(|1\rangle \langle 0| \hat{b} + \hat{b}^\dagger |0\rangle \langle 1| \right).\end{aligned}\quad (9)$$

It is interesting to note that this Hamiltonian provides a physical picture of the origin of entanglement generated between modes b and c . If mode a starts in a coherent state, then the interaction with the atom imprints a phase from mode a onto the probability amplitude for energy level 2, so that there is a coherence induced between levels 0 and 2. This coherence is transferred to modes b and c on transition to the atomic ground state so as to produce a nonvanishing expectation value $\langle \hat{b}\hat{c} \rangle$ even though $\langle \hat{b} \rangle = 0 = \langle \hat{c} \rangle$, which is a result of the entanglement between the modes. We may view this as a manifestation of interference between the two possible excitation pathways between the states $|0\rangle$ and $|2\rangle$ [22–27].

We can recover our initial nonlinear optical Hamiltonian by applying perturbation theory, as used in microscopic derivations of the nonlinear susceptibility [12]. Here we adopt a slightly different approach, in which we derive an effective Hamiltonian [28, 29] for the three interacting field modes based on time-independent perturbation theory to obtain, in operator form, the shift to the atomic ground-state energy. This is a simple extension of the treatments given in many quantum-mechanics textbooks [30–33], although we require perturbation theory up to third order, which is one order higher than that found in most texts. For these reasons, it is worth pausing to present a few details of the calculation. To order zero in the fields, our perturbed atomic ground-state energy is

$$\hat{H}^{(0)} = \langle 0 | \hat{H}_F + \hat{H}_A | 0 \rangle = \omega_a \hat{a}^\dagger \hat{a} + \omega_b \hat{b}^\dagger \hat{b} + \omega_c \hat{c}^\dagger \hat{c} + \mathcal{E}_0. \quad (10)$$

The first-order correction is zero,

$$\hat{H}^{(1)} = \langle 0 | \hat{V} | 0 \rangle = 0. \quad (11)$$

At the second order, we find a shift of the energies of modes a and b ,

$$\hat{H}^{(2)} = \frac{\langle 0 | \hat{V} | 1 \rangle \langle 1 | \hat{V} | 0 \rangle}{\mathcal{E}_0 - (\mathcal{E}_1 - \omega_b)} + \frac{\langle 0 | \hat{V} | 2 \rangle \langle 2 | \hat{V} | 0 \rangle}{\mathcal{E}_0 - (\mathcal{E}_2 - \omega_a)} = -\frac{g_a^2}{\delta} \hat{a}^\dagger \hat{a} - \frac{g_b^2}{\Delta} \hat{b}^\dagger \hat{b}. \quad (12)$$

Finally, at the third order, we find a term coupling the three modes,

$$\hat{H}^{(3)} = \frac{\langle 0 | \hat{V} | 2 \rangle \langle 2 | \hat{V} | 1 \rangle \langle 1 | \hat{V} | 0 \rangle + \langle 0 | \hat{V} | 1 \rangle \langle 1 | \hat{V} | 2 \rangle \langle 2 | \hat{V} | 0 \rangle}{[\mathcal{E}_0 - (\mathcal{E}_2 - \omega_a)][\mathcal{E}_0 - (\mathcal{E}_1 - \omega_b)]} = \frac{g_a g_b g_c}{\delta \Delta} \left(\hat{a}^\dagger \hat{b} \hat{c} + \hat{b}^\dagger \hat{c} \hat{a} \right). \quad (13)$$

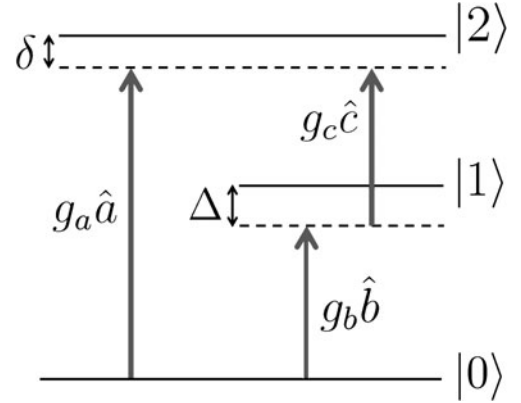


Fig. 1. Electronic energy level scheme and couplings for a microscopic theory of our nonlinear optical process. Here, the detunings δ and Δ are $\delta = \mathcal{E}_2 - \mathcal{E}_0 - \omega_a$ and $\Delta = \mathcal{E}_1 - \mathcal{E}_0 - \omega_b$, where \mathcal{E}_0 , \mathcal{E}_1 , and \mathcal{E}_2 are the energies of the states $|0\rangle$, $|1\rangle$, and $|2\rangle$, respectively.

If we combine these together and drop the unimportant constant ground-state energy, \mathcal{E}_0 , then we arrive at an effective Hamiltonian for the three interacting fields,

$$\hat{H} = \omega_a \hat{a}^\dagger \hat{a} + \omega_b \hat{b}^\dagger \hat{b} + \omega_c \hat{c}^\dagger \hat{c} - \frac{g_a^2}{\delta} \hat{a}^\dagger \hat{a} - \frac{g_b^2}{\Delta} \hat{b}^\dagger \hat{b} + \frac{g_a g_b g_c}{\delta \Delta} (\hat{a}^\dagger \hat{b} \hat{c} + \hat{b}^\dagger \hat{c}^\dagger \hat{a}). \quad (14)$$

The fourth and fifth terms in this Hamiltonian correspond to Stark shifts of the ground state but appear here as a modification of frequencies of the cavity modes. These account for the effective refractive index associated with the presence of the atom, and we can include these in the frequencies for the modes, $\omega'_a = \omega_a - g_a^2/\delta$ and $\omega'_b = \omega_b - g_b^2/\Delta$, to give

$$\hat{H} = \omega'_a \hat{a}^\dagger \hat{a} + \omega'_b \hat{b}^\dagger \hat{b} + \omega_c \hat{c}^\dagger \hat{c} + \frac{g_a g_b g_c}{\delta \Delta} (\hat{a}^\dagger \hat{b} \hat{c} + \hat{b}^\dagger \hat{c}^\dagger \hat{a}), \quad (15)$$

which becomes our initial Hamiltonian, Eq. (1), if we set $\kappa = g_a g_b g_c / \delta \Delta$ and relabel our AC Stark-shifted shifted frequencies ω'_a and ω'_b as ω_a and ω_b . At a fundamental level, these frequency shifts appear because we have used fixed cavity modes in our analysis, with predetermined wavelengths. The relabeling has a physical interpretation, which is that we shift to modes with a small difference in wavelength such that the process is resonant with the shifted frequencies.

Our perturbative approximation has reproduced the Hamiltonian, which has the energy spectrum with no lower bound. To show that this is a consequence of the approximations used to derive it, we need only show that the microscopic Hamiltonian from which it was derived, Eq. (8), does not have this property. To this end, we evaluate the expectation value of our microscopic Hamiltonian for a general atomic state, $\sum_i c_i |i\rangle$, and the field coherent state $|\alpha\rangle_a |\beta\rangle_b |\gamma\rangle_c$. For this state, we find

$$\begin{aligned} \langle \hat{H} \rangle &= \omega_a |\alpha|^2 + \omega_b |\beta|^2 + \omega_c |\gamma|^2 + \sum_{i=0}^2 \mathcal{E}_i |c_i|^2 \\ &+ g_a (\alpha c_0 c_2^* + \alpha^* c_2 c_0^*) + g_c (\alpha c_1 c_2^* + \alpha^* c_2 c_1^*) + g_b (\alpha c_0 c_1^* + \alpha^* c_1 c_0^*). \end{aligned} \quad (16)$$

This expectation value tends to $+\infty$ as the amplitudes of the coherent states become large, and the problem of unbounded negative eigenvalues does not arise. We conclude that this unphysical behavior arises as a consequence of extending a perturbative theory beyond the bounds of its validity. It is not difficult to see where this breakdown occurs; if the couplings between the atomic levels become large compared with the detunings (for example, $g_a \sqrt{N_a} \gg \delta$), then we start to find a significant probability for the atom to be found in one its excited states, and this invalidates the atomic ground-state assumption used in deriving the approximate Hamiltonian, Eq. (15).

4. Conclusions

The problem we have identified with the simple Hamiltonian, Eq. (1), applies to a very wide range of such model Hamiltonians used to describe quantum effects in nonlinear optics, including those used with great success to describe the generation of entangled states. That we can continue to use these model Hamiltonians with confidence comes from the fact that the dynamics predicted by these models cannot enter the regime in which unphysical behavior would emerge. That this is true mathematically is ensured by the conservation of the quantities \hat{M}_1 and \hat{M}_2 given in Eq. (5), which mean, in turn, that if we ensure that the maximum photon number in our analysis is sufficiently small, then we can be

confident in the predictions made using our model Hamiltonian. If we do push the model towards higher photon numbers, then we will, at some stage, need to abandon our simple multimode interaction term and include, explicitly, the dynamics of the nonlinear medium.

Perhaps, we should, if only occasionally, acknowledge the fact that the Hamiltonians we use habitually in modeling nonlinear optical processes should be used with caution [1], and that they form a valid description only when operating in a restricted state space, in which the photon number is not too large.

End note

Stig Stenholm was a wonderful man, a brilliant and ingenious physicist, a scholar, a caring nurturer of young scientists and, perhaps above all, an inquisitive, far-seeing and deep thinker. We worked together for many years, albeit intermittently, and I owe to him far more than these few words can adequately express. Looking back, I find it surprising that we published only nine papers together [34–42], but these papers constitute only a very small part of the fields we explored, in physics, mathematics and, latterly, in philosophy, especially the philosophical foundations of quantum theory [43].

Let me conclude by quoting the final paragraph from Stig's last book, *The Quest for Reality* [44], in which he sought to reconcile distinct philosophical views of quantum theory. He wrote:

The situation is far from satisfactory, but it may, in the end, be the best world image available to our limited human intellect. If that is so, we have to be grateful for what we get. Chasing rainbows has never uncovered the treasures. But the display of colors is magnificent.

This sums up the man I knew rather well: “you never really finish a problem,” he once told me, but the fun is in the challenge to understand.

Acknowledgments

Let me put on record the debt of gratitude that I owe to Stig Stenholm, as mentor, inspiration, poser of challenging puzzles (like the one addressed here) and, most importantly, as a friend.

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References

1. S. Stenholm circa (1986) private communication. His statement, if I remember correctly, was “Does it bother you that your Hamiltonian has a spectrum that is unbounded from below?” The very same question was posed soon afterwards, also, by K. Rzażewski after the presentation of a conference paper.
2. W. H. Louisell, *Radiation and Noise in Quantum Electronics*, McGraw-Hill, New York (1964).
3. J. Peřina, *Quantum Statistics of Linear and Nonlinear Optical Phenomena*, Kluwer, Dordrecht (1991).
4. P. Meystre and D. F. Walls (Eds.), *Nonclassical Effects in Quantum Optics*, American Institute of Physics, New York (1991). This is a reprint collection of important early papers in quantum optics, many of them dealing with quantum nonlinear optics.
5. D. F. Walls and G. J. Milburn, *Quantum Optics*, Springer-Verlag, Berlin (1994).
6. L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics*, Cambridge University Press (1995).
7. M. O. Scully and M. S. Zubairy, *Quantum Optics*, Cambridge University Press (1997).
8. S. M. Barnett and P. M. Radmore, *Methods in Theoretical Quantum Optics*, Oxford University Press (1997).
9. H.-A. Bachor, *A Guide to Experiments in Quantum Optics*, Wiley-VCH, Weinheim (1998).
10. R. Loudon, *The Quantum Theory of Light*, 3rd ed., Oxford University Press (2000).

11. H. J. Carmichael, *Statistical Methods in Quantum Optics*, Springer, Berlin (2008), Vol. 2.
12. R. W. Boyd, *Nonlinear Optics*, 3rd ed., Elsevier, Amsterdam (2008).
13. G. New, *Introduction to Nonlinear Optics*, Cambridge University Press (2011).
14. R. Bonifacio and G. Preparata, *Lett. Nuovo Cimento*, **1**, 887 (1969).
15. D. F. Walls and R. Barakat, *Phys. Rev. A*, **1**, 446 (1970).
16. S. M. Barnett and P. L. Knight, *Opt. Acta*, **31**, 435 (1984); Erratum: **31** 1203 (1984).
17. A. Yariv, *Quantum Electronics*, 2nd ed., Wiley, New York (1975).
18. M. Schubert and B. Wilhelmi, *Nonlinear Optics and Quantum Electronics*, Wiley, New York (1986).
19. P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics*, Cambridge University Press (1990).
20. A. G. Newell and J. V. Moloney, *Nonlinear Optics*, Addison-Wesley, Redwood City, CA (1992).
21. P. Mandel, *Nonlinear Optics*, Wiley-VCH, Weinheim (2010).
22. S. P. Krinitzky and D. T. Pegg, *Phys. Rev. A*, **33**, 403 (1986).
23. S. J. Buckle, S. M. Barnett, P. L. Knight, et al., *J. Mod. Opt.*, **33**, 1129 (1986).
24. B. W. Shore, *The Theory of Coherent Atomic Excitation: Vol. 2, Multilevel Atoms and Incoherence*, Wiley, New York (1990).
25. Z. Ficek and S. Swain, *Quantum Interference and Coherence*, Springer, New York (2004).
26. S. Stenholm and K.-A. Suominen, *Quantum Approach to Informatics*, Wiley, Hoboken, NJ (2005).
27. B. W. Shore, *Manipulating Quantum Structures Using Laser Pulses*, Cambridge University Press (2011).
28. D. J. Klein, *J. Chem. Phys.*, **61**, 786 (1974).
29. C. E. Soliverez, *Phys. Rev. A*, **24**, 4 (1981).
30. D. Bohm, *Quantum Theory*, Prentice-Hall, New York (1951); reprinted by Dover, New York (1989).
31. R. H. Dicke and J. P. Wittke, *Introduction to Quantum Mechanics*, Addison Wesley, Reading, MA (1960).
32. E. Merzbacher, *Quantum Mechanics*, 2nd ed., Wiley, New York (1970).
33. L. D. Landau and E. M. Lifshitz, *Quantum Mechanics*, 3rd ed., Butterworth Heinemann, Oxford (1977).
34. M.-A. Dupertuis, S. M. Barnett, and S. Stenholm, *J. Opt. Soc. Am. B.*, **4**, 1102 (1987).
35. M.-A. Dupertuis, S. M. Barnett, and S. Stenholm, *J. Opt. Soc. Am. B.*, **4**, 1124 (1987).
36. C. R. Gilson, S. M. Barnett, and S. Stenholm, *J. Mod. Opt.*, **34**, 949 (1987).
37. S. M. Barnett, S. Stenholm, and D. T. Pegg, *Opt. Commun.*, **73**, 314 (1989).
38. S. M. Barnett and S. Stenholm, *J. Mod. Opt.*, **47**, 2869 (2000).
39. S. Franke-Arnold, E. Andersson, S. M. Barnett, and S. Stenholm, *Phys. Rev. A*, **63**, 052301 (2001).
40. S. M. Barnett and S. Stenholm, *Phys. Rev. A*, **64**, 033808 (2001).
41. J. Salo, S. M. Barnett, and S. Stenholm, *Opt. Commun.*, **259**, 772 (2006).
42. S. Croke, S. M. Barnett, and S. Stenholm, *Ann. Phys. (N. Y.)*, **323**, 893 (2008).
43. S. M. Barnett and S. Stenholm, "Causal interpretation of quantum mechanics: an alternative to Bohmian mechanics," Unpublished note (2009).
44. S. Stenholm, *The Quest for Reality*, Oxford University Press (2011).