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Coherent transients theorem: A comment

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Schenzle, Wong, and Brewer have recently proposed a theorem [Phys. Rev. A 22, 635 (1980)] which states that the coherent transient signals produced in an optically thin gaseous sample of atoms are limited to a certain time interval after the excitation process which produces them. We comment on the range of validity of this theorem.

Recently Schenzle, Wong, and Brewer[1] (SWB) described a theorem on coherent transients which may be paraphrased as follows: The coherent transients (e.g., free-induction decay and echoes), produced in an optically thin sample by a "generalized excitation pulse" of total duration T, may be delayed by at most a time T from the cessation of the excitation pulse. SWB prove this theorem in the case of single-frequency copropagating-traveling-wave excitation of samples of two-level atoms, and conjecture that the theorem may be extended to multifrequency copropagating-traveling-wave excitation of samples of (three or more)-level atoms. We find that there are certain subtle difficulties associated with the generalization of SWB's theorem, and would like to point out situations in which long-delayed[2] coherent transient signals may arise. We restrict our attention here to echo phenomena, although similar considerations will apply to other coherent transient effects.

As is well known, an echo[3,4] consists basically of the following phenomena: An initial excitation field[5] places atoms at each location in a superposition of their energy eigenstates. The phases of all atomic superposition states are initially uniform at each position, but their overall phase generally varies with position. When the atomic superpositions at a given location are in phase, a net electric-dipole moment exists,[6] and if we assume that the overall local phase varies with position in an appropriate manner,[7] the sample emits an intense burst of radiation. Because of atomic motion, the initial sample order decays, i.e., the atoms dephase with respect to each other. A subsequent excitation field (or fields) causes the relative phase evolution to reverse, leading to a sample which is, at a later time, again phase ordered and which consequently emits an echo. Evidently, *the time of the echo will be determined by the relative dephasing and rephasing rates of the atomic superpositions.*

In the well-known two-pulse photon echo,[4,8–10] the dephasing and rephasing rates are identical; consequently, the dephasing interval (the interval between the two excitation fields) equals the rephasing interval (the interval between the second excitation field and the echo). The echo occurs delayed from the second excitation field by a time T, where T is the interval between the two excitation fields. The theorem of SWB applies.

A long-delayed echo may occur if the dephasing rate of the echo-producing superposition is larger than its rephasing rate. There are two situations in which this can occur: (1) During the generalized excitation pulse, the echo-producing superposition is transferred from one pair of energy eigenstates to another pair (with one or no level in common). We call this the superposition-level-switching case. (2) Non-copropagating excitation fields within the generalized excitation pulse may, by virtue of the vector nature of the Doppler effect, change the phase evolution rate of a superposition associated with a given fixed pair of energy eigenstates. We call this the non-copropagating case. The first situation requires (three or more)-level atoms, but may arise in either gaseous or solid samples excited exclusively by copropagating-traveling-wave fields. The second situation may arise in (two or more)-level atoms, but is restricted to Doppler-broadened samples. We note that non-copropagating excitation fields were not discussed in SWB's original formulation of the transients theorem.

As an example in which a long-delayed echo

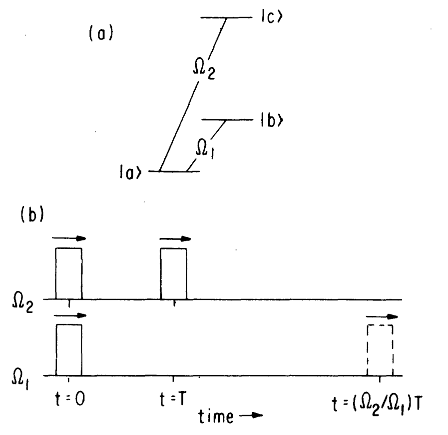

FIG. 1. The inverted-difference-frequency trilevel echo. In (a), the three energy levels and the two relevant transition frequencies are defined. In (b), the lower (upper) line gives the temporal sequence of excitation fields at frequency Ω_1 (Ω_2). The echo, represented by the dashed line, is of frequency Ω_1. As indicated by the arrows, all excitation fields and the echo copropagate.

may occur because of superposition-level switching, we consider the inverted-difference-frequency trilevel (IDF) echo.[11,12] This echo is produced by three copropagating-traveling-wave excitation fields in a three-level atomic system [see Fig. 1(a)]. The times and frequencies of the excitation fields are shown in Fig. 1(b). The excitation field(s) at frequency Ω_1 (Ω_2) have the wave vector $\vec{k}_1$ ($\vec{k}_2$), where $\vec{k}_1 \| \vec{k}_2$. An echo, represented by the dashed rectangle in Fig. 1(b) occurs at a time $[(\Omega_2/\Omega_1)-1]T$ after the final excitation field and has the wave vector $\vec{k}_1$. If $\Omega_2 > 2\Omega_1$, the echo occurs delayed by more than T from the last excitation field. Between the times $t=0$ and $t=T$ the IDF-echo-producing superposition is associated with the levels $|b>$ and $|c>$ and has a dephasing rate proportional to $|\vec{k}_2-\vec{k}_1|$. Since $\vec{k}_1 \| \vec{k}_2$, $|\vec{k}_2-\vec{k}_1| = (\Omega_2-\Omega_1)/c$, where we neglect dispersion throughout. After the last excitation field the superposition is associated with levels $|a>$ and $|b>$ and rephases at a rate proportional to $|\vec{k}_1+\vec{k}_2-\vec{k}_2| = |\vec{k}_1| = \Omega_1/c$. The change in phase evolution rate leads to the long echo delay.

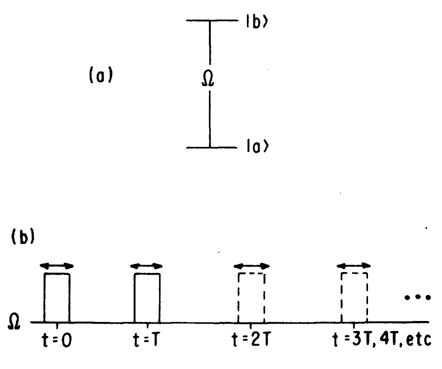

FIG. 2. Standing-wave photon echoes. In (a), we define the two-level system in which the echoes are generated. In (b), the solid lines represent standing-wave excitation fields and the dashed lines standing-wave echoes. Only two echoes are shown, but others (with varying amplitudes) occur for $t=4T$, $5T$, $6T$, ... All standing-wave fields are collinear.

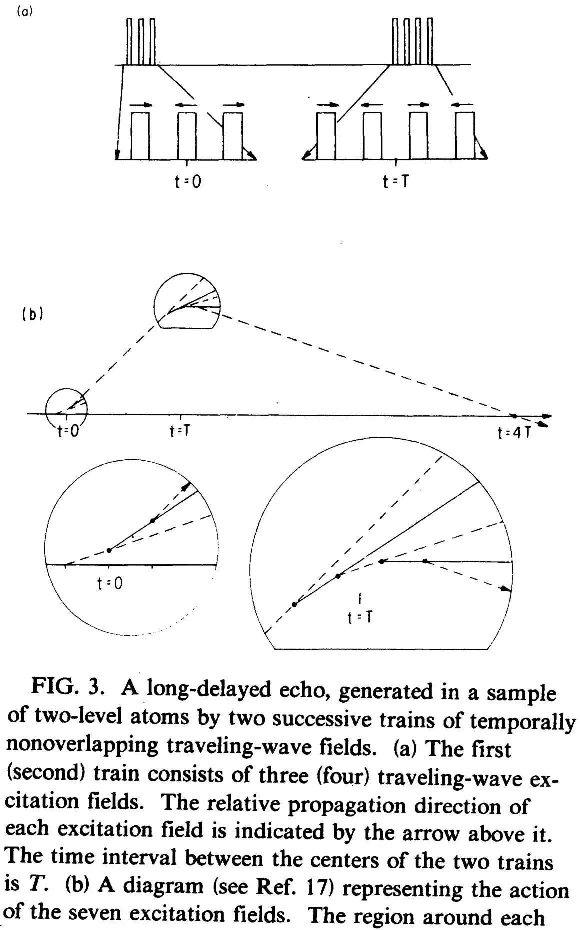

FIG. 3. A long-delayed echo, generated in a sample of two-level atoms by two successive trains of temporally nonoverlapping traveling-wave fields. (a) The first (second) train consists of three (four) traveling-wave excitation fields. The relative propagation direction of each excitation field is indicated by the arrow above it. The time interval between the centers of the two trains is T. (b) A diagram (see Ref. 17) representing the action of the seven excitation fields. The region around each train of excitation fields is shown in enlargements. The solid (dashed) lines represent phase information pertaining to the atom's ground state (excited state). Only the diagram lines relevant to the description of the echo which occurs at $t=4T$ and propagates along (←) are drawn. As explained in Ref. 17, the intersection of a dashed and solid line at $t=4T$ indicates the formation of the echo.

Note that if the excitation fields were not copropagating, the phase evolution rates would change as would then the time of the IDF echo. The IDF echo, generated by copropagating excitation fields, is correctly described by the modified coherent transients theorem presented by SWB in the following paper. Effects analogous to the IDF echo may also occur in solids (see, for example, Ref. 13).

Next consider the case of standing-wave photon echoes[14–16] which provide an example in which non-copropagating excitation fields lead to long-delayed echoes. A gaseous sample of two-level atoms irradiated by two successive collinear standing-wave fields separated by an interval T is found to emit echoes traveling in both senses along the excitation direction at multiples of T after excitation (see Fig. 2). Echoes have been observed delayed by $3T$ from the second standing-wave excitation field. These echoes are not echoes of echoes; rather, they arise because components of the superposition state established in the atoms by the first standing-wave excitation field have dephasing rates which are proportional not only to $|\vec{k}|$, where $\vec{k}$ is the wave vector associated with one traveling-wave component of each standing-wave excitation field, but also to multiples thereof. The second standing-wave field induces rephasing, but some components of the superposition rephase more slowly than they dephased.

Effects equivalent to standing-wave photon echoes would occur if each standing-wave field were decomposed into a series of alternately propagating traveling-wave fields which, while closely spaced (relative to T), do not overlap in time. In Fig. 3(a) we show the two-standing-wave fields replaced by three and four traveling-wave fields, respectively. In Fig. 3(b), we use the diagrammatic technique described elsewhere[17] to demonstrate the formation of a long-delayed echo. The vector nature of the Doppler effect, which makes long-delayed echoes possible in samples of two-level atoms, is also manifest in Doppler-free two- and three-photon spectroscopy.[18]

In summary, the original coherent transients theorem described[1] by SWB appears to be correct in the framework within which it was proven, i.e., copropagating-traveling-wave excitation of a sample of two-level atoms. Difficulties arise, however, when generalization to multifrequency or non-copropagating excitation fields is considered. *As long as only copropagating excitation is considered,* we know of no exceptions to SWB's modified coherent transients theorem which is presented in the following paper. Non-copropagating excitation, however, if carried to high enough order, can lead to arbitrarily long echo delay times.

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[2] We use this term "long-delayed" here to mean delayed by more than an interval T from the cessation of excitation.

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[5] The term "excitation field" is used to denote a distinct excitation pulse within the overall "generalized excitation pulse."

[6] Assuming of course, that the electric-dipole matrix element between the two energy eigenstates in superposition is nonzero.

[7] In the language of nonlinear optics, the sample must be "phase matched."

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