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Theorem on coherent transients: Response to a comment

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The limits of validity of our previous theorem on coherent transients are discussed with reference to a recent comment.

Mossberg and Hartmann[1] (MH) comment on a "theorem of coherent transients" developed by Schenzle, Wong, and Brewer[2] (SWB) citing examples which are not covered. Objection to the theorem's applicability arises when standing waves are used or when the excitation frequencies are different. In this comment, we further clarify the limits of validity of our theorem.

The theorem applies to nuclear magnetic resonance (NMR) and quantum optics, and is derived, within the rotating-wave approximation, for an optically thin atomic system subject to extreme inhomogeneous broadening and to coherent preparation by a pulsed electromagnetic field. The initial discussion of SWB considers a fundamental problem, the resonant excitation of a two-level quantum system by a single-frequency *traveling-wave* pulse of arbitrary shape, which is clearly defined in our Eq. (1).[2] The theorem states that when a pulse of finite duration T (interval $0 \leq t \leq T$) prepares a sample, the coherent emission which follows lasts only for an additional period T (interval $T \leq t \leq 2T$).

The validity of this theorem has already been demonstrated by experiments in the infrared[3] and radio frequency[4] regions, where the condition of strong inhomogeneous broadening is maintained. More recently, NMR measurements by Kunitomo *et al.*[5] confirm that the free decay signal for an inhomogeneously broadened transition terminates precisely at $t=2T$. Moreover, Kunitomo and Hashi[6] observe for the same inhomogeneously broadened system that spin-locked echoes, notched echoes, and new types of spin echoes are all confined to the time interval $T \leq t \leq 2T$. We emphasize that the theorem at this stage rests on the following assumptions: (1) extreme inhomogeneous broadening of a two-level quantum transition, (2) excitation by a single-frequency traveling-wave pulse of arbitrary shape, and (3) the rotating-wave approximation.

The subsequent discussion by SWB extends the theorem to a multilevel quantum system where the energy spacing, the number of fields, and their frequencies are arbitrary. However, we still assumed, though not explicitly stated, that all waves propagate collinearly in the forward direction as indicated in the formal structure of Eq. (13) of SWB. Any deviation from unidirectional excitation may result in spatial hole burning and may lead to the possibility of temporal rephasing of the spatial coherence created by such excitation. This kind of rephasing occurs in the standing-wave echo where the spatial coherence lasts for times $t \geq 2T$, as demonstrated both experimentally[7] and theoretically.[8] Obviously, the theorem does not apply in this case because the waves are no longer unidirectional.

Now consider the first example of MH, the inverted-difference frequency trilevel echo,[9] where the excitation is unidirectional, and the rephasing time satisfies $t > 2T$ for a suitable choice of frequencies. This example has caused us to realize that Eq. (13) of SWB is oversimplified and, thanks to their suggestion, we now introduce a modification. The difficulty lies in the definition of the tuning parameter

$$\Delta_{ij} = \omega_{ij} - \Omega_{ij} + kv_z ,$$

which asserts that the Doppler shift kv_z is the same for all transitions $i \rightarrow j$, making the SWB conclusion invalid for multifrequency excitation.

We now replace this definition by

$$\Delta_{ij} = \omega_{ij} - \Omega_{ij} + k_{ij}v_z, \quad k_{ij} \equiv \Omega_{ij}/c , \tag{1}$$

realizing that the density matrix has velocity-dependent components of the form $(k_{ij}v_z)$ for all i-j pairs. This change leads to a new result which is applicable when the excitation frequencies are different and also when multiphoton transitions occur.

The generalized theorem in its new form is derived in the same spirit as for the two-level case. A multilevel quantum system is assumed which is excited by a pulse of arbitrary shape consisting of traveling-wave components all propagating in the same direction with different frequencies. The rotating-wave approximation and the assumption of an optically thin sample are maintained in addition to that of strong inhomogeneous broadening. We further assume that for each n-photon i-j transition, there is at most one combination of n excitation fields which is resonant, as is usually the case in an experiment. This enables us to perform a generalized rotating-frame transformation by excluding any atomic structure which allows its energy levels to be coupled in more than one way. We denote the i-j transition frequency by ω_{ij} and the corresponding frequency of the resonant combination of excitation fields by Ω_{ij}. The quantities ω_{ij} and Ω_{ij} are elements of corresponding sets $\{\omega_{lm}\}$ and $\{\Omega_{lm}\}$, which embrace all possible transitions consistent with the fields applied. The coherent emission at time $t>T$ when averaged over the inhomogeneous line shape is given by

$$\langle\widetilde{\rho}_{ij}(t)\rangle=\frac{i}{2}\int_{-\infty}^{\infty}g(\Delta_{ij})d\Delta_{ij}\sum_{l}\int_{0}^{T}dt'e^{(i\Delta_{ij}-1/T_{ij})(t-t')}[\widetilde{\rho}_{il}(v_z,t')\chi_{lj}(t')-\chi_{il}(t')\widetilde{\rho}_{lj}(v_z,t')]\ , \tag{2}$$

where Δ_{ij} is defined by (1) and where we take $\widetilde{\rho}_{ij}(0)=0$.

The same analysis as in SWB follows. We have

$$\widetilde{\rho}_{il}(v_z,t')=\sum_{m}A_m e^{z_m t'}\ , \tag{3}$$

where z_m are the roots of the characteristic equation for the multilevel generalized Bloch equations. In the asymptotic limit $|v_z|\rightarrow\infty$, the roots assume the values

$$\lim_{|v_z|\rightarrow\infty} z_m=\text{const},\quad \pm ik_{ij}v_z\ ,$$

for all $i\neq j$ pairs. The constants are simply combinations of the population relaxation rates. Substitution of (3) into (2) followed by contour integration in the upper half plane results in the condition that for times

$$t_{ij}\geq(1+k_{\max}/k_{ij})T\ , \tag{4}$$

coherent emission for the i-j transition vanishes. Hence, *all* coherent emission should terminate for times

$$t\geq(1+\Omega_{\max}/\Omega_{\min})T\ , \tag{5}$$

where we now neglect dispersion, and $\Omega_{\max}$ and $\Omega_{\min}$ correspond to the maximum and minimum values of the set $\{\Omega_{lm}\}$. Note that (4) reduces to the two-level case, $t_{ij}\geq 2T$, when $\Omega_{\max}=\Omega_{ij}$.

Theoretical[9,10] predictions of trilevel echoes prepared by unidirectional excitation confirm the new result, Eq. (5). Furthermore, although Eq. (5) is valid only for unidirectional excitation, we note that the observed rephasing time of the sum-frequency trilevel echo,[11] in which counter-propagating waves are used, also occurs within the interval $T\leq t\leq(1+\Omega_{\max}/\Omega_{\min})T$. Theoretical[9,10] predictions of noncollinearly excited trilevel echoes also fall within the same interval.

Another exception to the theorem cited by MH is the multiple nuclear-spin echoes in solids observed by Solomon.[12] However, this spin system does not satisfy the essential requirement of being strongly inhomogeneously broadened, and therefore has no relevance.

In conclusion, we have delineated more explicitly the limits of validity of the theorem. The generalized form of the theorem states that an optically thin multilevel quantum system subject to a multifrequency radiation pulse of duration T and extreme inhomogeneous broadening, ceases to radiate coherent emission for times longer than $t=(1+\Omega_{\max}/\Omega_{\min})T$. This assumes the rotating-wave approximation and is valid only if all excitation fields propagate collinearly in the same direction and there exists at most one combination of excitation frequencies which is resonant with a particular transition. Within these restrictions, the number of fields, their frequencies, and the pulse shape remain arbitrary.

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