Quantum Beats in Photon Echo from Four-Wave Mixing

In a recent Letter, the issue of how to distinguish between the quantum beats (QB) and the polarization interference (PI) in four-wave mixing (FWM) experiments was raised [1]. For QB, it was argued that the FWM signal will have a maximum at $t_m = \tau + nT_B$, where n is an integer, τ is the time delay between the two short laser pulses, and T_B is the QB period; while for PI, the FWM signal would have a maximum at $t_m = 2\tau + nT_B$. Experiments exhibiting the two behaviors were reported and were attributed to QB and PI, respectively [1].

The theoretical analysis in the paper was based on a model with no dephasing or inhomogeneous broadening. However, the experimental results reported in [1] show that both time scales are comparable to T_B. The purpose of this Comment is the following: (1) We show that dephasing and inhomogeneous broadening can change qualitatively the outcome of the FWM signal, in terms of the relation between the signal peak times and the delay times. (2) Within the QB, by changing these physical parameters, one can find systems which cross over between the two behaviors [2]. Therefore t_m itself is not an unambiguous test to distinguish PI from inhomogeneously broadened QB; a more detailed analysis of the line shape is necessary.

We consider a three level system with energy levels 1, 2, and 3. Levels 1 and 2 lie close to each other, and are responsible for the QB. A third-order perturbation theory is used to calculate the FWM signal [2]. When both inhomogeneous broadening and dephasing are taken into account, we find the time-resolved FWM signal intensity is

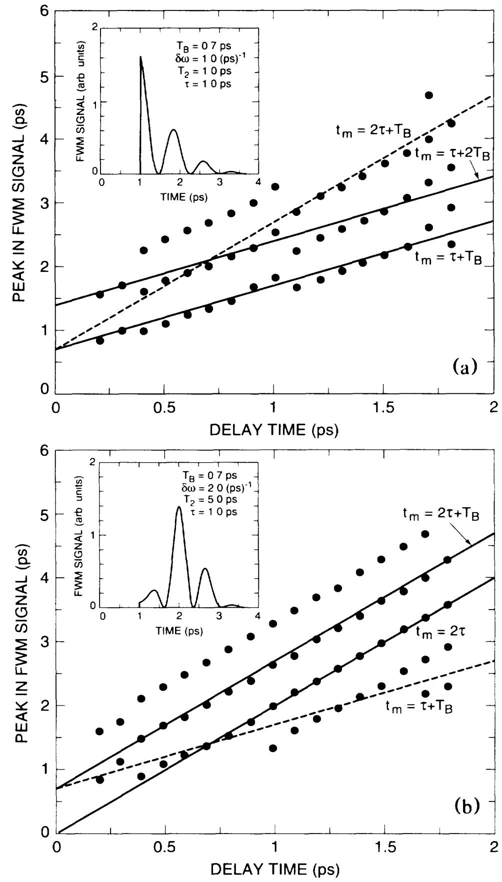

FIG. 1. The position of the peaks in the FWM signal as a function of time delay τ for (a) small inhomogeneous broadening and (b) large inhomogeneous broadening. Various parameters are given in the figures. The insets show the FWM temporal profile for $\tau = 1.0$ ps.

$$I^{\mathrm{FWM}}(t) = \alpha\theta(t-\tau)e^{-2t/T_2}\left[e^{-\frac{(t-2\tau)^2(\delta\omega)^2}{4}}\cos[\Delta\Omega(t/2-\tau)] + e^{-\frac{(t-\tau)^2(\delta\omega)^2}{4}}e^{-\frac{\tau^2(\delta\omega)^2}{4}}\cos(\Delta\Omega t/2)\right]^2, \quad (1)$$

where $\theta(t-\tau)$ is the step function, α is a constant related to the dipole matrix elements, T_2 is the dephasing time (assumed to be the same between levels 1 and 2, and level 3), $\delta\omega$ is the inhomogeneous broadening, $\Delta\Omega = 2\pi/T_B$ is the beating frequency, and τ is the time delay between the two pulses. When $\delta\omega$ and T_2 are included, the peaks in FWM will in general not occur at either $t_m = 2\tau + nT_B$ or $t_m = \tau + nT_B$. For a given $\delta\omega$, beyond a certain time delay τ, the peaks will always occur near $t = 2\tau + nT_B$. When the time delay $\tau = \pi/\Delta\Omega$, the two terms in Eq. (1) have opposite signs at any time t, making the overall FWM signal very small.

We have calculated using Eq. (1) the FWM peak position as a function of delay time between the pulses. In Figs. 1(a) and 1(b), we show our results obtained with parameters typical of the experiments like those in [1]. For relatively small inhomogeneous broadening [Fig. 1(a)], we find t_m and τ are basically related by $t_m = \tau + nT_B$ (n is an integer), with some noticeable deviations, especially at large τ which are better described by $t_m = 2\tau + nT_B$. This feature is also apparent in the experimental data in Fig. 2 in [1], but was left unexplained. For relatively large inhomogeneous broadening [Fig. 1(b)], we find that the two times are better related as $t_m = 2\tau + nT_B$, except for very small τ, as one would have expected from arguments given above.

Xuejun Zhu, Mark S. Hybertsen, and P. B. Littlewood
AT&T Bell Laboratories
Murray Hill, New Jersey 07974

Received 14 February 1994
PACS numbers: 42.50.Md, 73.20.Dx, 78.47.+p, 78.66.–w

[1] M. Koch *et al.*, Phys. Rev. Lett. **69**, 3631 (1992).
[2] Intuitively, even in the case of QB, with sufficient inhomogeneous broadening, the usual photon echo phase matching condition demands the FWM signal be maximum at $t_m = 2\tau$. Details of our results will be given in a future paper.

0031-9007/94/73(1)/209(1)$06.00