Interference of temporally nonoverlapping light pulses in degenerate four-wave mixing

É. A. Manykin, N. V. Znamenskiĭ, D. V. Marchenko, E. A. Petrenko, and M. A. Selifanov
Kurchatov Institute Russian Science Center, 123182, Moscow, Russia

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An interference of the signals of degenerate four-wave mixing excited coherently by two pairs of pulses which do not overlap in time has been observed experimentally for the first time. This interference arises from a superposition of two frequency population gratings which are produced within the nonuniform-broadening line of the working transition by two pairs of pulses.

Degenerate four-wave mixing (DFWM) with a time delay is generated by a train of three exciting pulses of identical frequency ω_L which are at resonance with some nonuniformly broadened transition $|2\rangle - |1\rangle$ of the atoms of a medium. The lengths of these pulses, as well as the time intervals t_{12} and t_{23} between them must be shorter than the time scale of the irreversible relaxation of the induced macroscopic polarization. The first two pulses (the "writing" pulses) give rise to a frequency-modulated population difference (a frequency population "grating") within the inhomogeneous-broadening line with a modulation period $2\pi/t_{12}$, in both the ground and excited states.[1] The diffraction of the third pulse (the "readout" pulse) by the frequency grating gives rise to a coherent response in the medium. This response is excited in the direction defined by the spatial matching conditiion $\mathbf{k} = -\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3$ (where $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are the wave vectors of the exciting pulses). It appears at a time t_{12} after the passage of the third pulse. One might ask how the response would change if the medium were excited by an additional pair of pulses before the application of the readout signal if the relative phase difference of these additional pulses could be varied smoothly with respect to the phase difference of the writing pair. Our purpose in the present study was to answer this question.

Let us examine in more detail the coherent resonant application of a pair of identical square pulses with an area[2] $\Theta_L \ll 1$, a length t_L, and a relative time separation t_{12} to a medium of inhomogeneously broadened two-level atoms with a width at half-maximum $\Delta\omega_{\text{in}}$. We assume that the spectral width $\Delta\omega_L$ of the pulses is smaller than the width of the inhomogeneous line ("narrow-band excitation"). In this case the following formula can be derived for the difference (n) between the populations in the upper level $|2\rangle$ and the lower one $|1\rangle$:

$$n = n^{(0)} + n^{(2)}, \tag{1}$$

where $n^{(0)} = -1$ is the difference in populations in the absence of excitation, and the increment $n^{(2)}$ is given by

$$n^{(2)} = -n^{(0)} \frac{1}{2} \left(\frac{\mu_{12}}{\hbar}\right)^2 \Theta_L^2 \frac{\sin^2\left[\frac{(\omega_L-\omega_0)t_L}{2}\right]}{\left[\frac{(\omega_L-\omega_0)t_L}{2}\right]^2} \times \cos^2\left[\frac{(\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}+(\omega_L-\omega_0)t_{12}+\omega_0 t_{12}}{2}\right]. \qquad (2)$$

Here ω_0 is the central frequency of the $|2\rangle - |1\rangle$ atomic transition, and μ_{12} is its matrix dipole element.

Expression (2) describes the appearance of a population grating inside the line of the nonuniformly broadened $|2\rangle - |1\rangle$ transition. It can be seen from this expression that, if the medium is excited by an additional pair of pulses before the readout signal is applied, then the frequency grating produced by this additional pair of pulses will interfere with the grating produced by the first pair, resulting in an amplification or weakening of the DFWM with a time delay. Under the assumption that the two gratings are formed independently in the medium (this assumption is always valid under the condition $\Theta_L \ll 1$), it follows that the mutual annihilation of these gratings occurs as the result of a superposition in the frequency domain, within a small quantity $\pi(\omega_L - \omega_0)/\omega_0$, if the time between the pulses of the additional pair satisfies $t'_{12} = t_{12} \pm \pi/\omega_0$. Figure 1 shows the results of a numerical calculation of the frequency

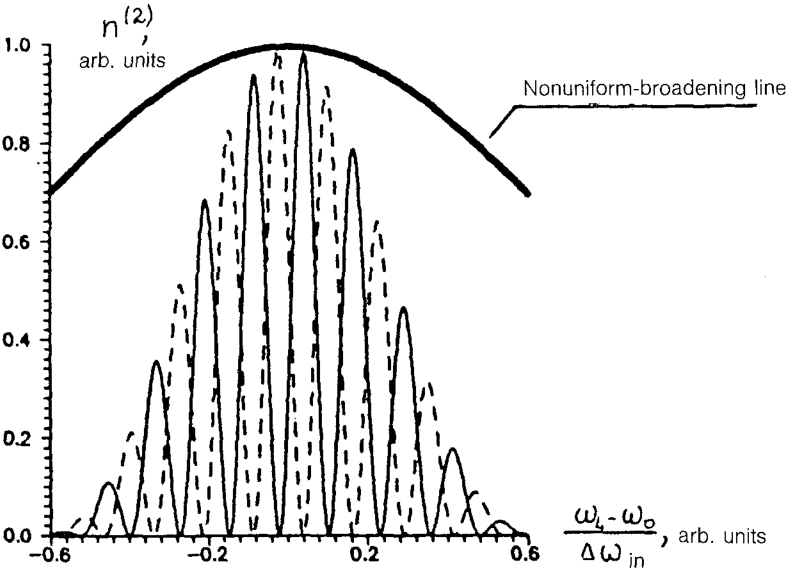

FIG. 1. Numerical calculation of the increment in the population difference between the upper and lower levels of a nonuniformly broadened ensemble of two-level atoms during coherent application of two pairs of pulses to the ensemble.

dependence of the increment in the populations of the lower and upper levels for the case in which two out-of-phase frequency gratings of populations are formed in a medium. It is particularly interesting to note that $n^{(2)}$ does not vanish in this case; i.e., the creation of the second grating in the working medium, out of phase with the first grating, does not result in a restoration to the original state, $|1\rangle$.

An interference of DFWM signals excited coherently by two pairs of pulses has been observed experimentally on the apparatus described in Ref. 3. The only modifications of the procedure were that the initial laser beam was split into five, rather than three, beams and that a train of five pump pulses of identical intensity was formed with the help of optical delay lines. The time interval between the pulses of the first pair was $t_{12} = 40 \pm 2$ ns, as before. The second (additional) pair was produced from the first pair and brought into coincidence with it by means of an optical system consisting of a combination of a Michelson interferometer and a delay line. This pair arrived at a time $t_{11'} = 100 \pm 2$ ns after the first pulse and had a delay $t_{1'2'} = t_{12} + \Delta$. The value of Δ could be varied highly precisely over a small interval by inserting a thin plane-parallel glass plate in one of the beams of the second pair. When this plate was rotated, the optical path length changed, so the time $t_{1'2'}$ changed. The readout pulse was applied 220 ns after the first pulse. The working medium was a LaF_3:Pr^{3+} crystal cooled to 4.5 K. The longitudinal and transverse relaxation times on the ${}^3H_4$–${}^3P_0$ transition of the Pr^{3+} ion are 47 μs and 300 ns, respectively, at these temperatures. In other words, the conditions for coherent excitation were satisfied well.

The optical layout which we used made it possible to spatially observe two DFWM signals with a time delay: signal 1, which arose from the first pair and the readout pulse (with the second pair blocked), and signal 1′, which arose from the second pair and the readout pulse (with the first pair blocked). These signals appeared at the same time (t_{12}), within the small quantity Δ, after the application of the readout signal to the medium. Frames *a* and *b* in Fig. 2 are photographs of the spatial distribution of intensity of these signals, respectively. Frame *c* is an interference pattern formed as a result of the superposition of the two echo signals 1 and 1′ upon the application of all five pump pulses to the working medium. When the glass plate was rotated, we observed a shift of the interference orders of this pattern. The appearance of this pattern, rather than the complete suppression of the echo response, is evidence that the wave vectors of the DFWM signals 1 and 1′ ($\mathbf{k}$ and $\mathbf{k}'$) are slightly different. The reasoning here is that, if the frequency gratings are produced in the medium by two pairs of pulses with wave vectors ($\mathbf{k}_1$, $\mathbf{k}_2$) and ($\mathbf{k}_1'$, $\mathbf{k}_2'$), which differ only slightly, then the diffraction by this grating of the readout pulse with the wave vector $\mathbf{k}_3$ should lead to the formation of two coherent DFWM signals, propagating in the directions

$$\mathbf{k} = -\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3, \qquad \mathbf{k}' = -\mathbf{k}_1' + \mathbf{k}_2' + \mathbf{k}_3'. \tag{4}$$

The superposition of these signals gives rise to the interference pattern with a spatial period $1/|\mathbf{k} - \mathbf{k}'|$ on the screen. From the frequency of the interference fringes we can estimate the angle between the vectors $\mathbf{k}$ and $\mathbf{k}'$. For our conditions, this angle turns out to be $\simeq 0.5$ mrad, with a laser-beam divergence $\simeq 1$ mrad. We attribute the

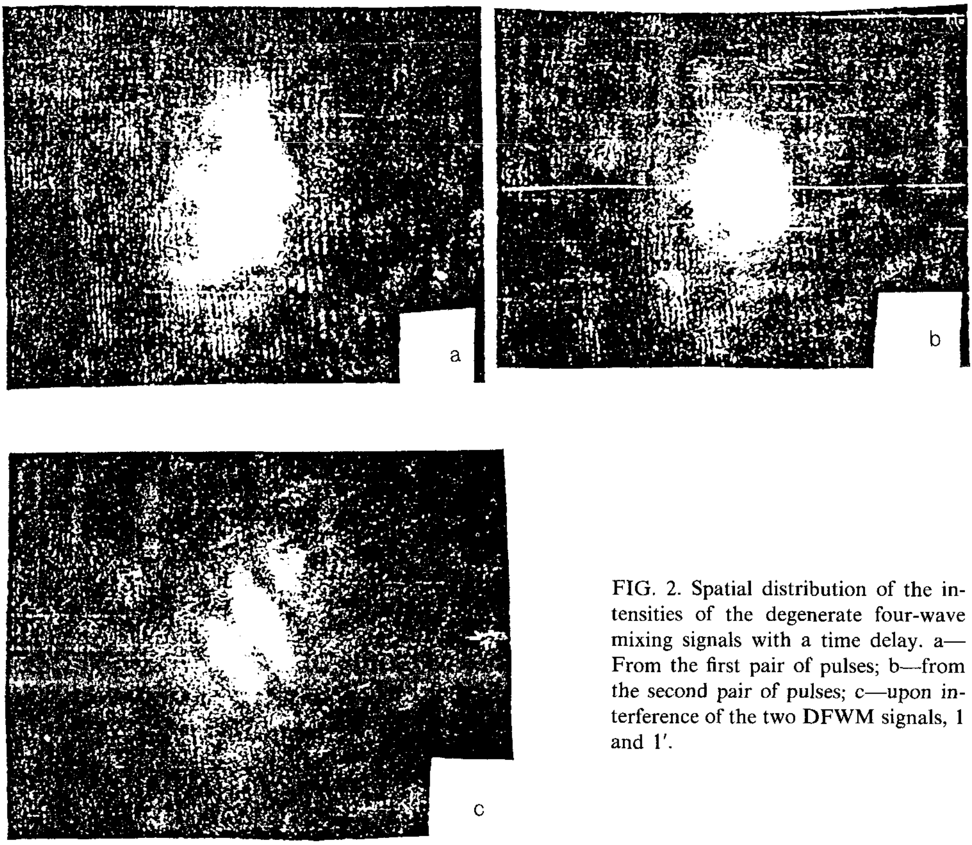

FIG. 2. Spatial distribution of the intensities of the degenerate four-wave mixing signals with a time delay. a—From the first pair of pulses; b—from the second pair of pulses; c—upon interference of the two DFWM signals, 1 and 1′.

circumstance that the vectors **k** and **k**′ could not be brought into coincidence exactly in these experiments to the large difference between the arms of the Michelson interferometer (30 m), which was necessary for the time separation of the writing and auxiliary pairs of pulses. Nevertheless, these results demonstrate that it is possible in principle to suppress (or amplify) DFWM with a time delay by means of an auxiliary pair of pulses, whose phase difference can be varied smoothly with respect to the phase difference of the pair of writing pulses.

[1] K. Duppen and D. A. Wiersma, Opt. Soc. Am. **3**, 614 (1986).
[2] A. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms,* Wiley-Interscience, New York, 1975.
[3] É. A. Manykin, N. V. Znamenskiĭ, D. V. Marchenko *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **54**, 172 (1991) [JETP Lett. **54**, 168 (1991)].

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