Subfemtosecond beats in an interference of the fields of Rayleigh scattering and four-wave mixing

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Subfemtosecond optical beats have been observed during resonant three-wave mixing of ultrashort laser pulses in a dye solution. This effect is a consequence of an interference of the Rayleigh scattering field with the field generated in the four-wave mixing.

DeBeer *et al.*[1] have reported the observation of subfemtosecond optical beats in four-wave mixing in Na vapor of the beams from two lasers at resonance with different D lines. Under the experimental conditions of Ref. 1 these beats were a consequence of the formation of two light-induced gratings. In the present experiments we have observed subfemtosecond beats for signals generated during three-wave mixing in a resonant medium of the pulses from a single laser, and in this case the beats have a different source: an interference of the Rayleigh scattering field and the four-wave-mixing field.

The experimental arrangement is shown in Fig. 1. Pulses from a dye (rhodamine

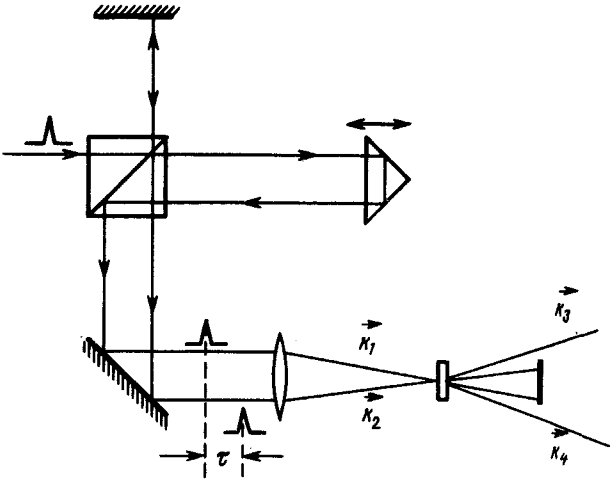

FIG. 1. Experimental arrangement.

6 G) jet laser with composite mode locking (an active locking through synchronous pumping with pulses from an argon laser and a passive locking by means of a nonlinear absorber) with a length ≈ 1 ps (estimated from autocorrelation measurements by the method of noncollinear second-harmonic generation) entered a Michelson interferometer. The interferometer was designed in such a way that two spatially separate pulses could be obtained, and the time delay (τ) between these pulses varied slowly (at 1 fs/s). When the two pulses were focused at an angle $\approx 1°$ into a cell holding a dye solution (DODS1 in ethylene glycol, concentration of 10^{-3}M, layer thickness of 100 μm), we observed signals of a resonant three-wave mixing along the directions $\mathbf{k}_3 = 2\mathbf{k}_2 - \mathbf{k}_1$ and $\mathbf{k}_4 = 2\mathbf{k}_1 - \mathbf{k}_2$ (k_j, where $j = 1, 2$, are the wave vectors of the excitation pulses). These signals were detected by photodetectors. Figure 2 shows the energy of these signals versus the delay time.

It was found that the length of the envelopes of the observed signals corresponds to the coherence time estimated from the width of the generation spectrum, not the duration of the laser pulses. For the upper envelope there is typically a single intensity maximum, while for the lower one there are two intensity minima (Fig. 2a). The period of the modulation on the slopes of the envelopes is equal to the period of the optical oscillations in the laser light (1.95 fs at a generation frequency $\omega = 17\,100$ cm^{-1}; Fig. 2b). At short delay times we observe a modulation near the signal maxima with a period half as long, of subfemtosecond length (Fig. 2c). For the Rayleigh-scattering signals detected at directions different from $\mathbf{k}_3$ and $\mathbf{k}_4$, the modulation period corresponds to the period of the optical oscillations.

To interpret the results, let us analyze the scattering which is excited by the laser pulses:

$$\mathbf{E}(\mathbf{R},t) = \frac{1}{2}\sum_{j=1,2}\{\mathbf{e}_j\mathcal{E}(t - \mathbf{n}_j\mathbf{R}/v - \tau_j)\exp[-i(\omega_j(t-\tau_j) - \mathbf{k}_j\mathbf{R})] + \text{c.c.}\}\,. \quad (1)$$

The scattering field in the directions k_n ($n = 3,4$) for an observation point far from

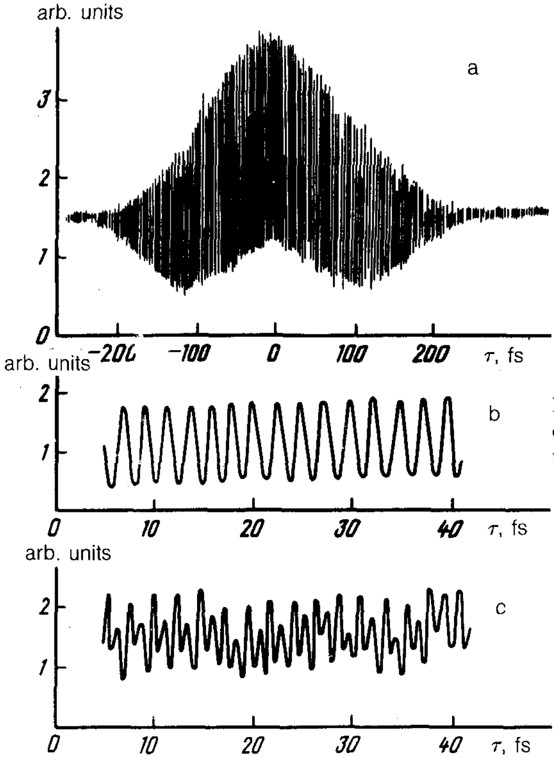

FIG. 2. Dependence of the scattering signals on the delay. a—Envelopes of signals; b—femtosecond beats; c—subfemtosecond beats.

the source, at a radius vector $\mathbf{R}_0$, can be written as follows, where we are incorporating the Rayleigh scattering and the four-wave mixing:

$$\mathbf{E}'_{\mathbf{k}_n}(\mathbf{R}_0, t) = \mathbf{E}_\mathrm{R}(\mathbf{R}_0, t) + \mathbf{E}_{\mathbf{k}_n}(\mathbf{R}_0, t), \tag{2}$$

where, according to Ref. 2,

$$\mathbf{E}_{\mathbf{k}_n}(\mathbf{R}_0, t) \sim \int_V d^3\mathbf{r}$$

$$\times \exp[-i\omega(t-(2\delta_{4n}-\delta_{3n})\tau)+i\mathbf{k}_n\mathbf{r}]\,\mathcal{E}(T-\delta_{4n}\tau)\int_0^\infty dt'\,\mathcal{E}(T-t'-\delta_{4n}\tau)$$

$$\times\ \mathcal{E}^*(T-t')-\tau\delta_{3n}) \equiv A_{4\mathbf{p}_n}(\tau)\exp[-i\omega(t-(2\delta_{4n}-\delta_{3n})\tau)] \tag{3}$$

is the field from the four-wave mixing, which is determined by an integral over the volume of the scattering medium (V), $\tau_1 = 0$, $\tau_2 \equiv \tau$, $T = t - (\mathbf{R}_0 - \mathbf{r})\mathbf{n}/v(\mathbf{n}_1 \approx \mathbf{n}_2 \approx \mathbf{n}^3 \approx \mathbf{n}_4 = \mathbf{n})$, $\delta_{jj'}$ is the Kronecker delta, and

$$\mathbf{E}_{\rm R}(\mathbf{R}_0,t) = \frac{\omega^2}{8\pi c^2 R_0}$$

$$\times \sum_{j=1,2} \left(\int_V d^3\mathbf{r}(\alpha \mathbf{e}_j)\mathscr{E}(T_{\rm R}-\tau_j)\exp\left[-i\omega\left(t-\frac{R_0-(\mathbf{n}_{\rm R}-\mathbf{n}_j)\mathbf{r}}{v}-\tau_j\right)\right]\right)_\perp$$

$$\equiv \exp(-i\omega t)[A_{\rm R_1}+A_{\rm R_2}\exp(i\omega\tau)] \tag{4}$$

is the Rayleigh-scattering field. Here also, $T_{\rm R} = t - [(R_0 - (\mathbf{n}_{\rm R} - \mathbf{n})\mathbf{r}]/v)$; α is the scattering tensor; and $(...)_\perp$ means the projection of the corresponding vector perpendicular to $\mathbf{k}_{\rm R}$. Expression (4) was derived through a generalization of the analysis of Ref. 3 to the time-dependent case.

The energy of the light scattered in the direction k_n is $J_{\mathbf{k}_n} \sim \int_{-\infty}^{\infty} dt\,\langle|\mathbf{E}'_{\mathbf{k}_n}|^2\rangle$, where the angle brackets mean all of the necessary averagings. The τ dependence of $J_{\mathbf{k}_n}$ is determined by the dependence of $|\mathbf{E}'_{\mathbf{k}_n}|^2$ on this parameter. Substituting (3) and (4) into (2), we find the following expression for $|\mathbf{E}'_{\mathbf{k}_n}|^2$:

$$|\mathbf{E}'_{\mathbf{k}_n}|^2 = |A_{\rm R_1}|^2 + |A_{\rm R_2}|^2 + \rho^2|A_{4{\rm R}_n}(\tau)|^2 + 2{\rm Re}[A_{\rm R_1}A^*_{\rm R_2}\exp(-i\omega\tau)]$$

$$+ 2{\rm Re}\{\rho\exp[i\omega\tau(2\delta_{4n}-\delta_{3n})](A^*_{\rm R_1}+A^*_{\rm R_2}\exp(-i\omega\tau))A_{4{\rm R}_n}/(\tau)\} \tag{5}$$

where ρ is a proportionality factor. The term in (5) which is independent of τ stems from the Rayleigh scattering and varies slowly over the time scale τ (of the four-wave mixing); the term which oscillates at the frequency ω is due to both Rayleigh scattering and an interference of Rayleigh scattering with the result of the four-wave mixing; and the term which oscillates at 2ω is caused exclusively by the interference of the Rayleigh scattering with the results of the four-wave mixing. This picture of the scattering field completely explains the observed features in the modulation and shape of the envelopes of the signals. In particular, the presence of two minima on the lower envelope can be explained on the basis that the term caused by the four-wave mixing is determined by a function of sixth power in the strength of the field which is acting, so it falls off more rapidly with increasing delay than the oscillatory terms associated with the interference (which are functions of the fields raised to the second and fourth powers).

These results are pertinent to an analysis of the shape and phase modulation of the pulses, since they can provide additional information about the interferometric method[4] based on second-harmonic generation. The reason is that in our case the signals are obtained in an interaction order higher than that in Ref. 4, and an increase in the interaction order should be accompanied by a strengthening of the dependence of the signals which are generated on the amplitude and phase characteristics of the pulses. A promising approach here would be to detect the four-wave-mixing signals in a situation in which pulses containing delayed and prompt components of the field are organized in both directions, $\mathbf{k}_1$ and $\mathbf{k}_2$. In this case the τ dependence of the four-wave-mixing signals would be given by

$$|\mathbf{E}_{\mathbf{k}_n}(\tau)|^2 \sim |\mathcal{E}(t) + \mathcal{E}(t-\tau)$$

$$\times \exp(i\omega\tau)|^2 \left[\int_0^\infty dt' |\mathcal{E}(t-t') + \mathcal{E}(t-t-\tau)\exp(i\omega\tau)|^2\right]^2. \tag{6}$$

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