# Phase Correlation of Laser Waves with Arbitrary Frequency Spacing

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The theoretically predicted correlation of laser phase fluctuations in Λ-type interaction schemes is experimentally demonstrated. We show that the mechanism of correlation in a Λ scheme is restricted to high-frequency noise components, whereas in a double-Λ scheme, due to the laser phase locking in a closed-loop interaction, it extends to all noise frequencies. In this case the correlation is weakly sensitive to coherence losses. Thus the double-Λ scheme can be used to correlate electromagnetic fields with carrier frequency differences beyond the GHz regime.

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The study of quantum interference effects in optical dense media, such as electromagnetically induced transparency (EIT) [1], is one of the most challenging fields in modern quantum optics research. In ideal EIT atoms are decoupled from resonant light fields and trapped into a dark state, which depends on the radiation amplitudes and phases. Perfectly phase correlated laser fields, i.e., fields with matched Fourier components, even though resonant, are not absorbed. In this Letter we show that in Λ-type excitation under the terms of EIT phase noise of one laser field is transferred to another one in a way that perfect phase correlation, i.e., $\omega_2 - \omega_1 = \text{const.}$, is given for the two laser fields $\omega_1$ and $\omega_2$. Such a perfect correlation is essential for high resolution in quantum interference applications. Among the basic correlation processes of field parameters in coherently prepared media are pulse matching [2], amplitude and phase matching [3], matched photon statistics [4], and intensity [5] and phase noise correlation [6,7], which extends also to quantized fields, including squeezing [8] and quantum entanglement [9]. In experiments phase correlated laser waves are usually produced via sideband modulation techniques [electro-optical, acousto-optical modulators, vertical cavity surface emitting lasers (VCSELs)] [10] or optical phase locking [11]. Hence the accessible frequency differences of phase correlated laser fields are restricted to the electronically available frequency limits, presently of the order of GHz. We show that any pair of laser frequencies, even with frequency spacing far beyond the GHz range, can be correlated in phase in the EIT regime. It can be done easily with a simple experimental setup, provided there is a suitable atomic or molecular medium whose energy level system allows us to combine resonantly the two frequencies in form of a Λ-type excitation scheme.

The Λ and closed double-Λ transition schemes are formed within the hyperfine sublevels of the Na $D_1$ line (590 nm) using $\sigma^+$-polarized and $\sigma^-$-polarized beams of two frequency stabilized cw-dye lasers (Figs. 1 and 2). To form a Λ scheme an acousto-optical modulator (AM1), driven at 1771.6 MHz, produces the first negative order modulation sideband $\omega_2$. An electro-optical phase modulator (PM), which causes a phase shift of 16.3 mrad/V, is used to modulate the phase of a part of $\omega_1$ by a 100 MHz band limited white frequency noise. For investigations in double-Λ configuration a second pair of frequency components ($\omega_3$, $\omega_4$) is generated by means of an electro-optical modulator (EM) driven at 885.8 MHz. The two first order modulation sidebands of carrier $\omega_0$ (Laser two is stabilized to the Λ-crossover resonance $3^2S_{1/2}$, $F = 1, 2$–$3^2P_{1/2}, F = 2$) match the Na ground state hyperfine splitting. To avoid an additional absorption background the carrier frequency $\omega_0$ is suppressed by an electronically stabilized Fabry-Perot etalon. It is combined with a polarizing beam splitter and a quarter wave plate to circumvent intense retroreflection into the dye laser. A defined phase relation between frequency pairs ($\omega_1$, $\omega_2$) and ($\omega_3$, $\omega_4$) is ensured by an internal frequency synchronization of the two frequency generators driving AM1 and EM [12]. The frequency pairs are prepared in circular polarization and transmitted collinearly through the absorption cell at typical incident intensities of 200 mW/cm$^2$ per frequency component. The corresponding Rabi frequencies $\Omega_j = d_j E_j/\hbar$ (with the dipole transition moments $d_j$) of the excited transitions $\omega_j$ are in the range of 10–30 MHz × 2π. After the cell the frequency pairs are separated by a quarter wave plate and a polarizer and observed separately with a 15 GHz InGaAs Schottky

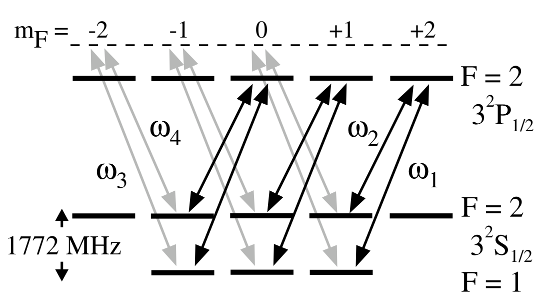


FIG. 1. Transition scheme within the Na $D_1$ line for Λ (black) and double-Λ excitation (black and gray).

 

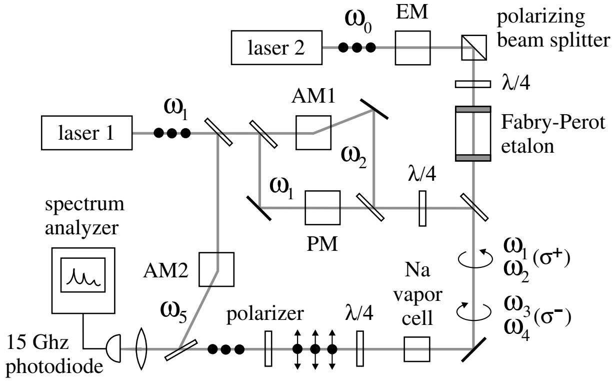


FIG. 2. Experimental setup. EM: electro-optical modulator, AM1/2: acousto-optical modulator, PM: phase modulator.

photodiode using heterodyne spectroscopy. The vapor cell is a 1 $\mathrm{cm}^3$ cube with a sidearm containing the sodium reservoir. The medium optical density $\tau(T)$ is controlled via this reservoir temperature $T$, which is stabilized with an accuracy of 1 °C. The windows are kept at higher temperature to avoid darkening. $\tau(T)$ was calibrated via an absorption measurement on transition $3^2S_{1/2}, F = 2$–$3^2P_{1/2}, F = 2$. The cell is placed inside of three mutually orthogonal Helmholtz coils to compensate stray magnetic fields. To observe the spectral noise distribution $S_i(\tau, \omega)$, i.e., the intensity spectrum of phase noise of a single laser component with carrier frequency $\omega_i$, a part of $\omega_1$ is shifted by 260 MHz using acousto-optical modulator AM2 ($\omega_5 = \omega_1 + 260$ MHz) and superposed with the transmitted $\omega_i$ laser beam. As $\omega_5$ is free of noise, the spectrum $S_i(\tau, \omega)$ can be observed directly by taking the beat signal $S_{i5}$ at frequency $|\omega_i - \omega_5|$ via a 2.8 GHz electronic spectrum analyzer.

A theoretical analysis of the correlation of phase fluctuations for EIT in $\Lambda$ systems is performed in Refs. [6,7]. It is shown that the spectrum $W_\psi$ of phase-difference fluctuations $\delta\psi = \delta\varphi_1 - \delta\varphi_2$ of two laser fields $\omega_1$ and $\omega_2$ decays with the propagation distance: $W_\psi(z, \omega) = W_\psi(0, \omega)\exp[-\int_0^z 2\kappa(z', \omega)dz']$. Here $z$ is the propagation distance, and $\omega$ is the noise frequency. Contributions from the atomic noise are neglected, which is justified under the conditions of EIT. Obviously the laser phase fluctuations $\delta\varphi_1$ and $\delta\varphi_2$ become more and more correlated with the propagation distance. Slight extension of the theory in [6,7], assuming equal dipole moments, decay rates and Rabi frequencies of the involved atomic transitions and no phase correlation before interaction, gives for the individual spectra of phase fluctuations $W_{11}$, $W_{22}$ and for the cross-correlation $W_{12}$ the following approximate dependencies on the propagation path

$$W_{11}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 + e^{-2x}) + \frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega)), \quad (1)$$

$$W_{22}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 + e^{-2x}) - \frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega)), \quad (2)$$

$$\mathrm{Re}W_{12}(z, \omega) = \frac{1}{4}(W_{11}(0, \omega) + W_{22}(0, \omega))(1 - e^{-2x}), \quad (3)$$

$$\mathrm{Im}W_{12}(z, \omega) = \frac{1}{2}(W_{11}(0, \omega) - W_{22}(0, \omega))e^{-x} \quad (4)$$

with $x = \int_0^z \kappa(z', \omega)dz'$. These equations show that two laser fields transfer and exchange their noise properties in the course of propagation. After a sufficiently long propagation path, the noise spectra of both fields are identical and the fields are perfectly correlated: $|2W_{12}| = \sqrt{W_{11}W_{22}}$ [13]. If initially only one of the fields has fluctuations above shot noise ($W_{11}(0, \omega) \neq 0, W_{22}(0, \omega) = 0$), then we observe from the above equations the noise transfer from component $\omega_1$ to the initially noise free component $\omega_2$: $W_{22}(z, \omega) = \frac{1}{4}W_{11}(0, \omega)(1 - e^{-x})^2$. Simultaneously the cross correlations $\mathrm{Re}W_{12}(z, \omega)$ grow exponentially at comparable rate.

In order to test this prediction experimentally, we observe the intensity noise spectra $S_1(\tau, \omega)$ and $S_2(\tau, \omega)$ by taking the beat signals $S_{15}$ and $S_{25}$, respectively (the centers of the noise spectra are normalized to zero frequency by subtracting the frequency difference of the two carriers). Further we observe the dependence of the FWHM $\Delta S_{12}$ of the beat signal $S_{12}$ on $\tau$. An increasing optical path length $z$ is simulated by changing the optical density $\tau$ via cell heating. Usually the random frequency jitter of light emitted by two different lasers is uncorrelated, and the width $\Delta S_{12}$ of the beat signal at frequency difference $\omega_2 - \omega_1$ is the sum of the linewidths of the two lasers. Phase correlation due to EIT improves with $\tau$, thus $\Delta S_{12}(\tau)$, similar to $W_\psi(z)$, reflects the degree of correlation between different frequency sidebands of $\omega_1$

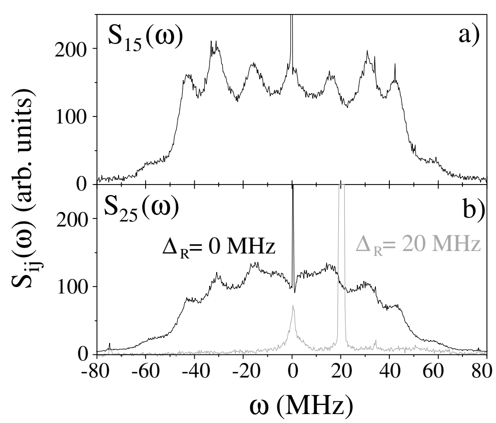


FIG. 3. Phase noise transfer at $\tau = 7.3$: Intensity spectra $S_{15}(\omega)$ (a) and $S_{25}(\omega)$ (b), representing the spectra of phase fluctuations $S_1(\tau, \omega)$ and $S_2(\tau, \omega)$ respectively; Raman detuning $\Delta_R = 0$ (black curve) and $\Delta_R = 20$ MHz (gray curve).

and $\omega_2$. $\Delta S_{12} = 0$ corresponds to perfect correlation. Figure 3(a) shows a measurement of the beat signal $S_{15}$, which represents the spectrum of phase fluctuations $S_1(\tau, \omega)$ modulated onto carrier $\omega_1$. The wavelike structure is caused by the high-frequency noise amplifier characteristic, the central peak occurs at the exact carrier frequency difference. The measurement of beat signal $S_{25}$ [Fig. 3(b)—black curve] exactly represents the spectrum of phase fluctuations $S_2(\tau, \omega)$: A noise transfer from frequency component $\omega_1$ to the initially noise free frequency component $\omega_2$, as predicted by Eq. (4), is obvious.

Under the same assumptions as for Eqs. (1)–(4) we derived the noise transfer rate

$$\kappa = \kappa_0 \frac{\Gamma_g^2}{\Gamma_g^2 + (\omega - \Delta_R)^2} \frac{\omega^2}{\Gamma_g^2 + \omega^2}, \qquad (5)$$

where $\kappa_0 = 4\mu|\Omega|^2 N/\gamma^2\Gamma_g$ is the maximum rate, with transition coupling elements $\mu_j = \omega_j d_j^2/\hbar c$, spontaneous decay rates $\gamma_j$, Rabi frequencies $\Omega_j$ (all assumed equal), atom density $N$, Raman detuning $\Delta_R = \omega_1 - \omega_2 - \omega_{12}$, and the transparency window width $\Gamma_g = \Gamma + 2|\Omega|^2/\gamma$. $\Gamma$ is the dark state coherence decay rate. The noise transfer rate $\kappa$ shows a typical Lorentzian profile with respect to the Raman detuning $\Delta_R$, with the width $\Gamma_g$. Only for a small band of Raman detuning $\Delta_R$ around the noise frequency $\omega$ the transfer rate is of considerable magnitude—thus the efficient noise transfer is due to EIT. This fact is supported by our measurements: In Fig. 3(b), the second (gray) curve is obtained for $\Delta_R$ larger than the transparency window width. We see, that no efficient noise transfer occurs, except for the beat frequencies $\omega \approx \Delta_R = 20$ MHz, in correspondence with (5). We have performed such measurements for series of different Raman detunings and observed, that the noise transfer efficiency depends on $\Delta_R$ as a Lorentzian function, in very good agreement with Eq. (5).

An important feature follows from the factor $\omega^2/(\Gamma_g^2 + \omega^2)$ in Eq. (5): There is no fluctuations correlation for frequencies inside the transparency window $\omega < \Gamma_g$, independent on the values of $\Delta_R$! This is due to the adiabatic regime in this noise frequency range, where small variations in the laser phase are so slow, that the atom follows the evolution of the fields and remains in a dark state. Only high-frequency noise components $\omega > \Gamma_g$ are correlated. As the laser intensity itself exponentially decreases with optical density, and $\Gamma_g \sim \Omega^2$, the ultimate low-frequency threshold for phase correlation is determined by $\Gamma$. This fact is clearly demonstrated in our measurements: We observed the $S_{12}$ beat signal at different optical densities $\tau$ for zero Raman detuning. Since noise transfer happens, the spectra $S_1(\tau, \omega)$ and $S_2(\tau, \omega)$ are almost identical, and the corresponding beat signal $S_{12}$ shows a Lorentzian profile with a half width $\Delta S_{12}$ limited by $\Gamma_g$. The FWHM $\Delta S_{12}(\tau)$ was evaluated and depicted in Fig. 4(a): As $\omega_1$ and $\omega_2$ propagate, more and more lower noise frequencies $\omega$ become correlated, and $\Delta S_{12}(\tau)$ exponentially decreases and asymptotically approaches the limit set by $\Gamma$. The curve $\Delta S_{12}(\tau)$ was fitted by an exponential decay function yielding a dark state coherence decay rate $\Gamma = 0.3(1)$ MHz, which is in good agreement with measurements of $\Gamma$ [14] made independently of the present experiment. Additional measurements of $\Delta S_{12}(\tau)$ at $\Delta_R = 8$ MHz show that outside the EIT regime there is no correlation effect at all!

In principle, the rate $\Gamma$ can be made very small if the low lying states $|1\rangle$ and $|2\rangle$ of a $\Lambda$ system are close in energy. However, for a considerable energy difference of $\omega_1$ and $\omega_2$ (e.g., in the optical range), $\Gamma$ will be quite large, of the order of $\gamma$. Moreover, the correlation mechanism requires EIT, which in turn requires sufficient high intensities $|\Omega|^2 \gg \Gamma\gamma$, so that the transparency window gets even wider. Thus, in such realistic cases, only small parts of the phase noise spectrum can be correlated. This problem can be avoided if one uses the double-$\Lambda$ scheme: Here the phase noise spectra have approximately the same dependence on the propagation length as above—Eqs. (1)–(4)—but similar relations are valid for any pair of the four participating frequency components. Thus noise transfer and correlation of phase fluctuations occur among all four radiation fields. Essential for the double-$\Lambda$ system is the different noise transfer coefficient, which for the stationary situation is given by

$$\kappa \simeq \kappa_0 \frac{\Gamma_g^2 \cos\varphi_0 + (\omega - \Delta_R)^2(1 + \cos\varphi_0)}{\Gamma_g^2 + (\omega - \Delta_R)^2}. \qquad (6)$$

Here $\kappa_0$ is the same as in Eq. (5), and $\Gamma_g = \Gamma + 4|\Omega|^2/\gamma$ for a double-$\Lambda$ system. $\varphi_0 = (\varphi_1 - \varphi_2) - (\varphi_3 - \varphi_4)$ is the value of the mean relative phase of the transition excitation loop [3] at a given optical length. The phase

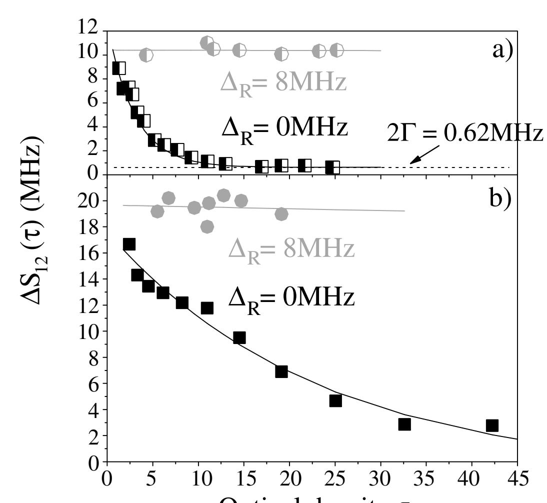


FIG. 4. FWHM $\Delta S_{12}(\tau)$ of the $\omega_1/\omega_2$ beat signal at 0 and 8 MHz Raman detuning: (a) In the $\Lambda$ scheme the exponential decay is limited by $\Gamma$; (b) In the double-$\Lambda$ scheme the exponential decay asymptotically approaches zero.

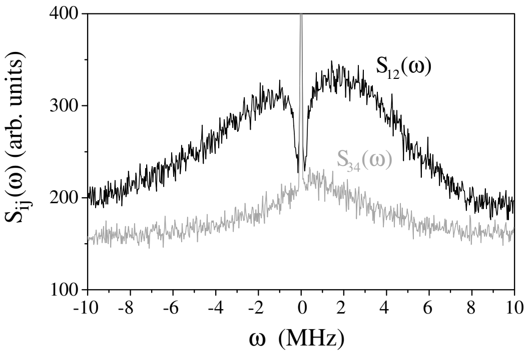


FIG. 5. Intensity spectra $S_{12}(\omega)$ and $S_{34}(\omega)$ taken separately at $\tau = 7.3$ in case of double-$\Lambda$ excitation.

$\varphi_0$ itself evolves with the propagation distance, and inside the transparency window $(\omega - \Delta_R) < \Gamma_g$ the phase $\varphi_0$ rapidly (with the rate $\sim 4\kappa_0$) approaches the value $2\pi n$ [3], while outside the transparency window it changes very slowly, and on a scale $\kappa_0^{-1}$ $\varphi_0$ is almost constant. For noise frequencies inside the transparency window $\kappa \simeq \kappa_0 \cos\varphi_0$, while outside $\kappa \simeq \kappa_0(1 + \cos\varphi_0)$. Consequently the noise transfer coefficient is not zero in the whole frequency range. In contrast to EIT in the $\Lambda$ system, the correlation of phase fluctuations takes place for all noise frequencies $\omega$, including the low-frequency range! This unlimited correlation is demonstrated in the experiment: After transmission in double-$\Lambda$ excitation the beat signals $S_{12}(\omega)$ and $S_{34}(\omega)$ (the latter is initially free of noise) are observed separately (Fig. 5): Because of the noise amplifier's cutoff frequency of 0.5 MHz the phase noise spectrum $S_1(\tau, \omega)$ and accordingly the beat signal $S_{12}(\omega)$ show a distinct dip around zero peak. Such a pronounced dip is not found in the $S_{34}(\omega)$ intensity spectrum, which confirms a phase noise transfer without frequency limits for the double-$\Lambda$ regime. Analogous to the measurements in Fig. 4(a) we evaluated the FWHM $\Delta S_{12}(\tau)$ [Fig. 4(b)]. In accordance with our model $\Delta S_{12}(\tau)$ exponentially decreases and (within the margin of fitting error) asymptotically approaches zero.

In double-$\Lambda$ excitation the noise transfer coefficient $\kappa \neq 0$ almost independent on the laser intensity, also for large $\Gamma$ (e.g., for large carrier frequency differences). Inside the transparency window the correlation happens, in general, slower than in the $\Lambda$ system, as can be seen from the different decay rates of $\Delta S_{12}(\tau)$ in Fig. 4. This is the price to pay for low-frequency noise correlation.

The double-$\Lambda$ system can be applied for a phase correlation of lasers with substantially different wavelengths (e.g., correlation of UV with IR) up to the shot noise (and even beyond—using entanglement [15]). In practice a double-$\Lambda$ excitation scheme is established easily—each of the two fields of an appropriate $\Lambda$ system can be shifted in frequency by an equal amount using sideband modulation, and afterwards all four resulting fields are superimposed in the medium. The process also works in the degenerated double-$\Lambda$ configuration [16]. Here the mean relative phase $\varphi_0$ is constant, and can easily be controlled and put to zero [17]. Such a setup might be relevant for any EIT application to modern nonlinear optics, where standard phase correlation techniques do not suffice. Besides high precision spectroscopy we expect promising applications in quantum information processing [18], quantum state engineering [19], or long distance quantum communication [20]. As to the realization of a quantum repeater, quantum correlated photon pairs have already been generated [21] using the EIT-based technology of light pulse storage [22]. Phase correlated excitation of optical materials with high nonlinearities and low loss might well become essential for the controlled generation of entangled states and quantum logic operations in future quantum computer design.

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