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Dynamical studies of non-degenerate two-wave mixing

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ABSTRACT

We experimentally study non-degenerate two-wave mixing with a harmonically varying frequency shift in dark ruby and colloidal media and compare the results to theoretical models for the detailed process in these two very different physical systems. Excellent agreement is found for both the radiative decay in ruby as well as the particle diffusion driven relaxation of nano-particle colloidal media. The results show that there is a universal asymmetry in the oscillating energy exchange waveform which arises from symmetry-broken grating dynamics of the in-phase and out-of-phase gratings.

1. Introduction

Non-degenerate two-wave mixing (NDTWM) process have permeated a number of fields and applications from photorefractive response and image correction to artificial Kerr media such as colloids. Colloidal systems have extremely low-intensity nonlinearity of the Kerr-type, which is driven by the local density changes which through an effective-medium model result in local index of refraction changes proportional to the intensity. The theory of NDTWM has been developed from the vantage point of a generic nonlinear index medium with a generic relaxation mechanism [1]. Various studies of NDTWM focusing on relaxation of ions in solids under varying conditions of temperature and electrical and magnetic field bias have been performed [2–8] as well as biological media such as bacteriorhodopsin [9] and in composite media comprised of polymer doped quantum dots [10,11]. In addition, more subtle processes such as pressure induced effects in sodium vapor and nematic liquid crystal films have been studied [12,13]. Colloidal media comprised of nanoscale particles in a liquid have been recently studied extensively with respect to and shown to exhibit a number of nonlinear optical phenomena, such as confinement and guiding of light in silver nanoparticles in acetone [14], tunable (including negative values) polarizability of synthetic colloidal suspensions of polytetrafluoroethylene nanoparticles in a glycerin–water solution [15], nonlinear coupling of light in colloidal gold nanosuspensions in water [16], as well as interactions between optical beams [17,18]. With regards to NDTWM in colloids, where relaxation of the formed particle density gratings is due to diffusion, a number of experiments have been performed with the primary focus being the elucidation of the NDTWM gain mechanism and the use of the technique to determine fluid properties and melting dynamics of strongly interacting colloidal crystals [19–22].

In all the systems studied to date the equations governing the process have been compared to experiment in the steady state limit of a constant frequency detuning in time. In this work, we study the ruby and the colloidal systems under conditions of a frequency shift between the two beams which varies sinusoidally in time. The motivation for this is to determine if the dynamical response is well described by the models as well as to determine if a more rapid determination of the relaxation constant can be implemented using a few cycles of this process.

2. Experiment

The experimental set up used to make the measurements is shown in Fig. 1. Solid state lasers operating at 671 nm and 532 nm with measured coherence lengths of 3 mm and 3m were used as the sources for the two physical systems studied. The laser was split into two beams of equal powers and one beam was frequency shifted by reflection from an ultrathin mirror mounted on a PZT which could be driven in the traditional triangle wave mode used in the quasi-steady state experiments in previous work, or driven by a sinusoidal function which results in the frequency shift which varies harmonically in time. The modulation frequency of the PZT was in the range of 30 Hz–100 Hz and the applied peak to peak voltages were 0–10 V.

The system was computer controlled and the powers were selected using an electro-optic crystal and polarizers. When experiments using

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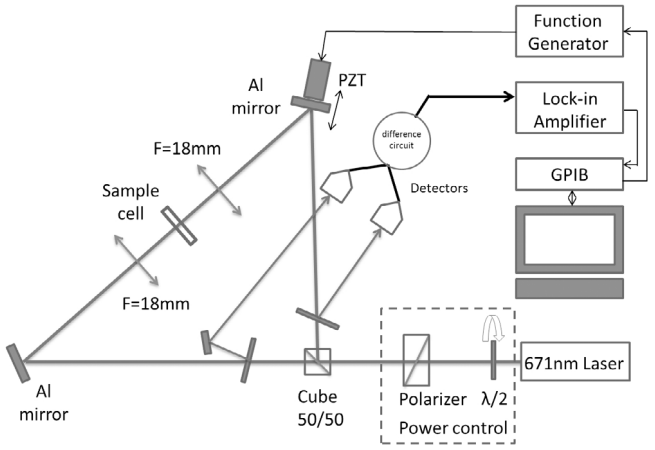

Fig. 1. Experimental setup for measuring NDTWM signal where two beams from a 671 nm laser (or 532 nm for ruby) with variable Doppler shift induced by the PZT are focused down to a spot diameter of 17 μm (13 μm) using lenses with 18 mm focal lengths.

the standard approach of triangle wave displacement to create square wave frequency shifts (periods of constant shift) was used, a lock-in amplifier was used to record the data at each voltage corresponding to a given frequency shift. At 37 Hz, the frequency shift using triangle modulation was shown to be 3546 Hz at a peak to peak voltage of 1 V. Typical scans of the shift versus NDTWM signal took approximately 30 min. In the case of the colloidal samples, where the viscosity of the liquid plays an important role, water was selected as the host fluid owing to its well-characterized temperature-dependent viscosity. The samples which were in 200 μm thick cuvettes were temperature controlled using a thermoelectric module and water cooling in a feedback loop. Spectroscopic measurements of the samples resulted in a single localized plasmon peak at 533 nm (expected for spherical particles) before and after the experiments were completed. This single peak is evidence [23] that no precipitation or flocculation took place during the experiments and is consistent with the diluted density of 10^{12} cm^{-3}.

3. Time dependent response of colloidal media

A colloidal medium comprised of water and gold nanospheres of diameter 50 nm with a coefficient of variation of 6% from Nanopartz Inc. was diluted to a particle density of 1×10^{12} cm^{-3} and placed in a 200 μm thick quartz cuvette and used as the sample for the experiments on sinusoidal modulation of the frequency difference of the two fields. The modulation is described as $\phi(t)$ which is given by:

$$\phi(t) = k\xi V_0 sin\,(2\pi f t) \tag{1}$$

where V_0 is the peak to peak amplitude of the two fields, k is the vacuum wavevector of the laser source in use, ξ is the displacement of the PZT per applied volt (2.77 μm/V) and f is the PZT drive frequency.

The equations governing the dynamical response of a colloidal medium in response to a moving intensity grating were given by McGraw and Rogovin [19] for the case of a gradient force acting on the particle. Describing the linearly polarized counter-propagating fields as:

$$\begin{aligned} E_1 &= E_0 e^{i(kx-\omega t+\phi(t))} \\ E_2 &= E_0 e^{i(-kx-\omega t)} \end{aligned} \tag{2}$$

results in a potential energy for a nanoparticle given by:

$$U = -g k_B T\,[1 + cos\,(2kx + \phi(t))] \tag{3}$$

where g is the normalized strength of the interaction energy relative to the thermal energy given by:

$$g = 8\pi n_h a^3\, Re\left\{\frac{m^2-1}{m^2+2}\right\}\frac{I_0}{c\,k_B T} \tag{4}$$

where $I_0 = \frac{1}{2}\epsilon_0 c n E_0^2$, $m = n_p/n_h$, n_p and n_h are the particle and host fluid refractive indices [24], and a is the particle radius. For 50 nm Au particles at 671 nm, $g = 0.05$ (input power of 40 mW focused to a diameter of 17 μm and resulting in a peak intensity of 13 kW/cm^2 at $T = 25$ °C. This field results in a traveling wave force which through the viscosity, μ, results in a velocity function in the limit of no inertial forces which is well justified. In addition, there are no polarization-dependent effects since the nanoparticles are spherical, the host medium is isotropic, and the two counterpropagating beams are polarized along the same direction.

The solutions for the density as a function of time and position in one dimension (x), are obtained using the ansatz:

$$\rho = \rho_0 + A(t)cos\,(2kx + \phi(t)) + B(t)sin\,(2kx + \phi(t))\,. \tag{5}$$

Following McGraw and Rogovin [19] and substituting into the Fokker–Planck equation and using the Einstein–Stokes relations between the diffusion coefficient results in the following equations for the amplitudes $A(t)$ and $B(t)$ for the in-phase and out of phase traveling grating components:

$$\dot{A}(t) + B(t)\dot{\phi}(t) + \frac{A}{\tau} = \rho_0 g\frac{1}{\tau} \tag{6}$$

$$\dot{B}(t) - A(t)\dot{\phi}(t) + \frac{B}{\tau} = 0. \tag{7}$$

The relaxation time τ in Eqs. (6) and (7) is given by $\tau = 4k^2n^2D$ which for the case of colloidal media is determined by diffusion, the mechanism by which the induced gratings relax. In all previous treatments, the $\dot{\phi}(t)$ term was set a constant equal to the frequency shift between two waves using a triangle to drive the PZT. In this limit the solutions are given by:

$$A = g\rho_0\frac{1}{1+(\delta\omega\tau)^2} \tag{8}$$

$$B = g\rho_0\frac{\delta\omega\tau}{1+(\delta\omega\tau)^2}. \tag{9}$$

The dependence of A and B on g shows that the colloidal system exhibits a Kerr medium response, where the population and the accompanying local index change are proportional to the intensity (E_0^2). As was shown in [19], this steady state population grating results in energy exchange between the two waves which is proportional to the out of phase grating amplitude, B. This is a general relationship which can be derived using an intensity dependent index of refraction which has an in phase and out of phase response relative to the intensity grating. Fig. 2 shows a fit of the result for $B(\delta\omega,\tau)$ and as is typically done, the relaxation time can be determined from the peak of the curve. In the case of a polydisperse colloidal particle size distribution, the fixed relation of the ratio of the peak to the full width at half maximum exceeds the value $2\sqrt{3}$. The contribution of electronic and thermal effects can be neglected on the basis of the measured relaxation time of 0.095 ms. The electronic response of gold is 9.4×10^{-15} s [24], and the thermal response of water is approximately 10^{-6} s [25] for grating decay at 671 nm wavelength.

When the harmonically varying frequency shift is used to excite the energy exchange, there is no quasi-steady state as in the triangle modulation case. Fig. 3 shows a typical result of a waveform obtained for the Au nanoparticle colloid driven with a 9.84 volt peak to peak displacement function on the PZT. As can be seen from the results, the dynamical theory of the process in colloids for the small $g \ll 1$ case provides a very accurate description of the system when the relaxation rate τ matches the value obtained from the quasi steady state results of Fig. 2. At a frequency of 37 Hz and a digital averaging of 520 scans, the data is recorded in less than 15 s. This data, which contains all the information required to determine τ is obtained in two orders of magnitude less time than with triangle wave modulation and phase sensitive detection.