New nonlinear Bragg diffraction devices

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Colloidal particles in solution are often charged due to ionization of acidic or basic functional groups attached to the particle surfaces during their synthesis. These charged colloidal particles dispersed in media with high dielectric constants show electrostatic interactions over relatively large distances between particles. The distances over which the interactions are energetically significant are determined by the number of surface charges, the ionic species present in the medium, the temperature and the medium dielectric constant. Aqueous high particle density colloidal solutions (10^{13} sphere/cc) with low concentrations of ionic impurities show large electrostatic interactions over relatively large distances (ca 1 micron). If the colloidal particles have identical charge, the lowest energy for the system is where the particles form an array in a cubic lattice. Thus, the particles self assemble into an ordered array called a crystalline colloidal array.[1-4] At high concentrations and at sufficient low solution ionic strengths, the system will self assemble to form a macroscopic single crystal which occurs either as a face centered cubic or a body centered cubic crystal (Figure 1).

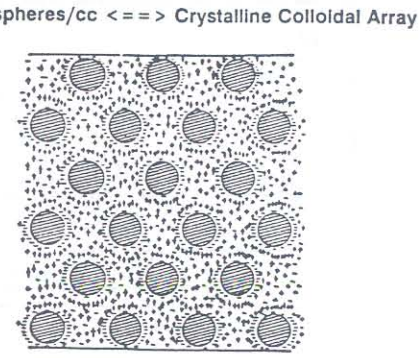

Figure 1. Crystalline colloidal array ordering.

Since the dielectric constant of the spheres, in general, differs from that of the medium, the periodic spacing of the spheres causes a periodic variation in the material dielectric constant. Electromagnetic radiation will Bragg diffract from this modulated dielectric constant, in a manner similar to the diffraction of x-rays from the modulated dielectric constant of atomic and molecular crystals.[1,5] A major difference occurs, however, in that the dielectric constant modulation for the crystalline colloidal array is much larger than that found for atomic and molecular crystals. Diffraction in the case of the colloidal crystalline arrays occurs in the dynamical diffraction limit as opposed to the kinematic limit for x-rays.[5]

We have extensively examined diffraction of light from these crystalline colloidal arrays and have quantitated the dependence of the diffraction properties on sphere size, medium refractive

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index, sphere charge, crystal ordering, lattice parameter and sphere interparticle interaction potential.[5-9] The diffraction of light almost follows Bragg's law and it is easy to prepare a single crystal of these colloidal arrays in which the longest set of planes are oriented parallel to a surface. In this case, Bragg diffraction will occur, for example, from the 110 plane of body centered cubic crystals or the 111 plane of face centered cubic crystals. The wavelength diffracted will change as a function of angle and closely follow Bragg's law as shown in Figures 2 and 3 which are adapted from reference 3. The diffraction efficiency is such that it is easy to decrease the transmittances of wavelength meeting the Bragg condition to less than 10^{-8} from colloidal crystals which are a few hundred microns in thickness. Light not meeting the Bragg condition freely transmits (T > 0.80). The development of optical devices using these colloidal crystalline arrays was first discussed in two of our patents,[4] and these devices were previously commercialized through licensing of the technology to EG&G Princeton Applied Research.

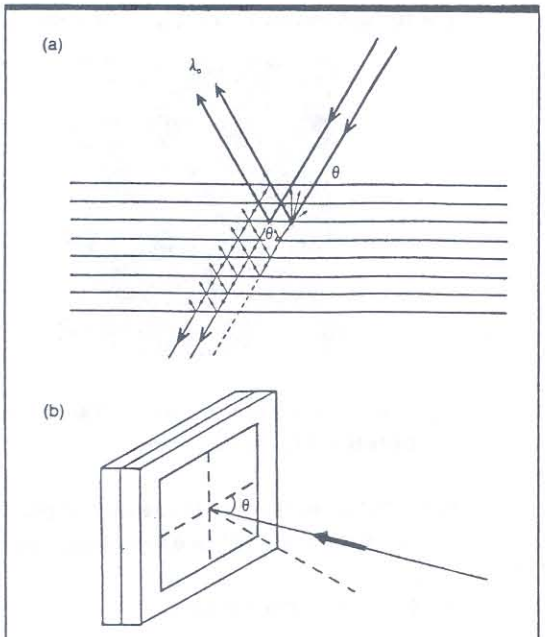

Figure 2. (a) Schematic representation of a set of layers in the colloidal crystal and the locus of incident Bragg glancing angles that fulfill the Bragg condition. (b) The orientation of a filter to the incident light beam.

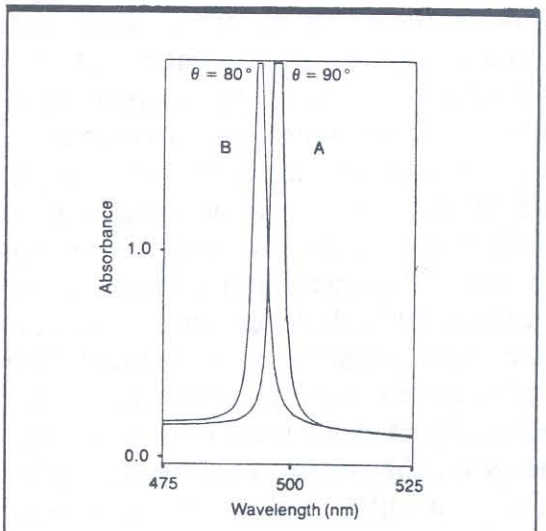

Figure 3. Spectra showing transmission through the Bragg diffraction filter as a function of filter orientation to the incident beam. At 90° incidence, the center rejection wavelength is 497 nm (A). If the filter is tilted to 80°, the rejected band shifts to 493 nm (B).

Our group has continued to develop these crystalline colloidal arrays and to utilize the unique self-assembling properties of the colloidal arrays to fix the dielectric constant periodicity in matter.[10] We are developing a nonlinear optical device which utilizes a nonlinear refractive index within the spheres or within the medium. The transmission of these devices would be a function of the incident light intensity. For example, at low light intensities, the refractive index of the spheres would be prepared to be identical to the refractive index of the medium. Thus, no dielectric constant modulation occurs and light freely transmits. However, since either the medium or the spheres has a nonlinear dependence of the refractive index on the incident light intensity, at high light intensities the refractive indices diverge from one another and the grating "pops up". Recently, a patent was issued to Spry[11] for such a device. Our challenge is to fabricate these crystalline colloidal arrays with nonlinear materials.

We have begun to develop a nonlinear colloidal array which utilizes a thermal nonlinearity. Figure 4 shows a colloidal sphere which has attached dyes that absorb radiation. In a theoretical examination of the thermal nonlinear response, we have calculated the change in the refractive index which would occur due to light illumination for a crystalline colloidal array of 83 nm spheres of polymethylmethacrylate dispersed in water in a body centered cubic crystal. This crystal is 400 microns thick and the colloidal spheres are refractive index matched to an aqueous solution containing high refractive index cosolvent which makes the refractive index of the solution equal to that of PMMA (n = 1.492). Under low light intensities, the crystal is completely transmittive, but for light meeting the Bragg condition incident on the crystal at the Bragg diffraction angle, the transmittance decreases as the spheres heat up. Kesavamoorthy et al.,[12] in a recent publication, calculated that the rate of temperature increase and relaxation for these spheres in water heated by a laser occurred in the nsec time frame.

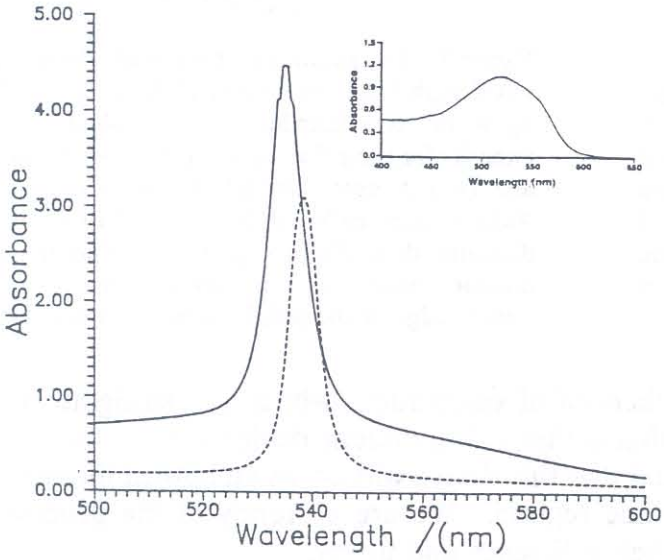

Figure 4. Visible absorption spectra of the large BCC colloidal crystals prepared from dyed (solid line) and undyed (dashed line) polystyrene spheres. The inset shows the visible absorption spectrum of Oil Red O in Ethyl Benzene (1.1×10^{-4} M). (from reference 6)

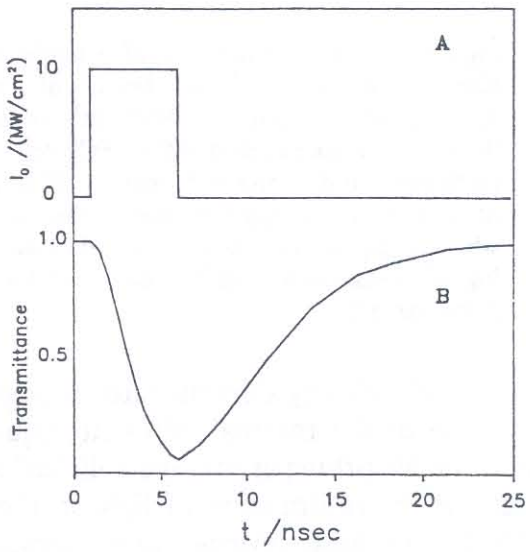

Figure 5. Time dependence of transmittance through a nonlinear optical switch. (A) Heat beam pulse shape. (B) Transmission of the probe beam of wavelength 514.5 nm incident at $\theta_B = 75°$ on an index-matched dyed colloidal crystal of 83 nm diameter PMMA particles as a function of time. (from reference 12)

Figure 5 shows the time dependence of transmittance through this crystalline colloidal array for an incident 5 nsec laser pulse which has a maximum intensity of 10 megaWatts/cm^2. Figure 5A shows the temporal intensity dependence of the incident laser beam while Figure 5B shows the time dependence of the transmittance of the crystal. Each sphere absorbs only 1% of the light falling on it. Within 5 nsec the refractive index of the spheres diverges from that medium sufficiently that less than 10% of the light transmits through the crystal. Also evident is that the system thermally recovers within ca 20 nsec. Figure 6 shows the response of the system for a longer incident pulse intensity. It is seen that the transmittance decreases to less than 1% within 10 nsec. The nonlinear response of the system is most clearly shown in Figure 7.

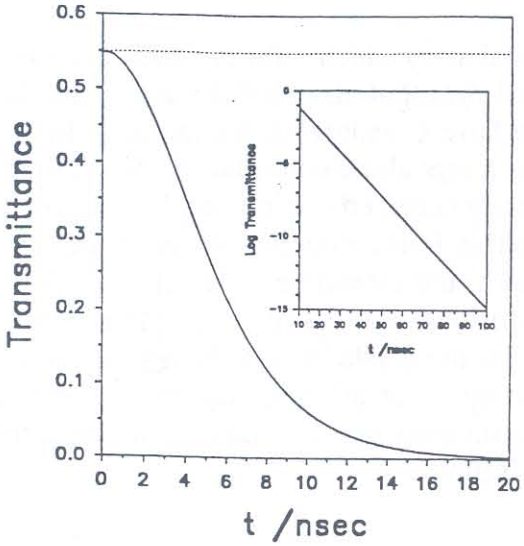

Figure 6. The transmittance of a nonlinear Optical Limiter of an incident beam of wavelength λ_h = 450 nm, of intensity 10 MW/cm^2 through a 50 μm thick dyed, index-matched PMMA FCC colloidal crystal (with particle diameter = 83 nm) at 90° as a function of time, (—) if Bragg condition is satisfied and (---) when Bragg condition is not satisfied. Inset shows the log of transmittance as a function of time. (from reference 12)

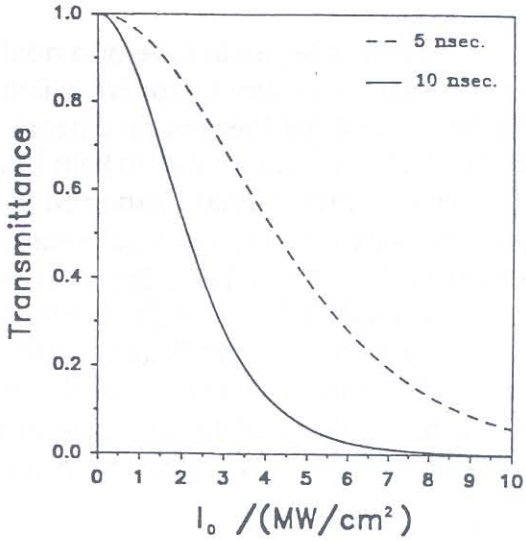

Figure 7. Transmittance of the probe beam of wavelength 514.5 nm incident at the Bragg angle θ_B = 75° as a function of the incident beam intensity for the pulse durations t_h of (–) 10 nsec and (---) 5 nsec, through an index-matched 400 μm thick colloidal BCC crystal of 83 nm diameter dyed PMMA particles. The transmittance plotted is that which occurs at the trailing edge of the pulse. (from reference 12)

We clearly demonstrate through this theoretical calculation, which is a straightforward exercise of the thermal diffusivity equation, that a thermal nonlinear device can be fabricated from an absorbing crystalline colloidal array and that this device will act as an optical limiter and can switch transmission of light in the nsec time regime. We are currently in the process of fabricating these devices and demonstrating their efficiency and utility.

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