# Modulation of the propagation speed of mechanical waves in silicon quantum dots embedded in a silicon-nitride film


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**Abstract:** Using a vectorial picosecond self-diffraction method, we evaluate the modification of the speed of the sound in a silicon-nitride film containing silicon quantum dots prepared by remote plasma-enhanced chemical vapor deposition. Our non-contact technique is based on the stimulation of the electrostriction contribution to the nonlinearity of index exhibited by the sample in a multiwave mixing laser experiment. We identified the electronic birefringence using two of the incident beams to generate a self-diffraction signal, then, we modified the third order nonlinear response by means of the optical Kerr effect given by a phase-mismatched third beam which induced electrostriction. Our results indicated that the speed of the sound in a silicon-nitride film can be simultaneously tailored by an electronic nonlinear refractive index, and by an electrostriction effect, both resulting from silicon quantum dots doping.





**OCIS codes:** (160.4330) Nonlinear optical materials; (190.3270) Kerr effect; (190.4223) Nonlinear wave mixing; (160.4236) Nanomaterials.


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## 1. Introduction

The propagation conditions related to waves in advanced materials is one of the most fascinating topics involved with nanoscale systems [1]. Currently, for instance, the most ordinary way for playing and recording sound is through electronic devices that day after day are becoming smaller, besides that the acoustic frequency generators nowadays are frequently assembled by means of compound systems and nanostructures [2]. A modulation in the propagation of a mechanical wave in an optical media can be achieved through intense optical signals by changes in the refractive index, and consequently, in density. Electrostriction is a common physical mechanism that originates a nonlinearity of refractive index by using high intensity Gaussian beams [3]; nevertheless it can be also obtained by sharp optical irradiations created by patterns of diffraction or interference [4]. Other different physical mechanisms that could give rise to an optical Kerr effect are the electronic polarization or an excited-state population, among others [5]. The manifestation of each one of these effects in nanostructures is strongly dependent on the wavelength and pulse duration of excitation [6]. The development of nanophotonic devices based on semiconductors, particularly, silicon quantum dots (Si-QDs), has attracted much the attention since apparently it gives the possibility of fabrication of low dimensional structures with potential applications in several areas where a strong and ultrafast optical response is required [7]. Different measurements of induced electrostriction, and manifestation of changes on acousto-electromagnetic interactions of sound in compound materials have been reported [8]. However, the ability of manufacturing new nonlinear optical materials with purposes associated with smart materials is still in progress [9]. It has been demonstrated that silicon nanocrystals exhibit an important nonlinear optical response [10]; and there is also a great interest for finding an enhancement in their optoelectronic properties for waveguiding [11], instrumentation of signals [12], luminescent properties [13], and mechanical features [14]. Furthermore, the study of the close relation between collective phenomena where optical and mechanical characteristics of silicon nanocomposites are involved looks to be motivating. Within this work, we investigate the modification of the speed of the sound in a silicon-nitride film containing Si-QDs. A non-contact technique based on vectorial self-diffraction and the stimulation of the electrostriction contribution to the nonlinearity of index given by phase-mismatched beams is presented.

## 2. Theory of picosecond vectorial self-diffraction enhanced by electrostriction

We consider two coherent pulses described as plane waves with the same frequency and different irradiances which interfere in a thin isotropic nonlinear medium. When a nonlinear refractive index is induced and an interference pattern with modulation of intensity or polarization results from this interaction, a self-diffraction response can be observed as the first order of diffraction travelling close to the neighborhood of the two incident beams. It has been demonstrated that comparing the self-diffraction effect originated by parallel and orthogonal polarization of the incident beams it is possible to measure the absorptive and

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refractive nonlinearities [15]. Since the electrostriction phenomenon takes some nanoseconds for its manifestation in the nonlinearity of refractive index [5], a picosecond self-diffracted pulse originated by a picosecond two-wave mixing is short enough in time for allowing the observation of an electrostriction effect responsible of a nonlinear refractive index. However, by using a longer excitation, or by previously inducing a high intensity laser irradiation into the sample, before the grating of index is generated by the two-wave mixing, then it is possible to measure the contribution of the electrostrictive force to the self-diffraction signals. With the purpose of separately identify the modification of the refractive index by electrostriction and by electronic nonlinearity, in this work we proposed to provide an additional brief third optical beam and a delay in the two-wave mixing for the self-diffraction experimental setup. We use a description of the field in terms of circular polarization components as it has been previously reported [15]. The amplitudes of the transmitted and self-diffracted waves are described by

$$E_{1\pm}(z)=\left[J_0\left(\Psi_{\pm}^{(1)}\right)E_{1\pm}+iJ_1\left(\Psi_{\pm}^{(1)}\right)E_{2\pm}\right]\exp\left(-i\Psi_{\pm}^{(0)}-\frac{\alpha(I)z}{2}\right), \tag{1}$$

$$E_{2\pm}(z)=\left[J_0\left(\Psi_{\pm}^{(1)}\right)E_{2\pm}-iJ_1\left(\Psi_{\pm}^{(1)}\right)E_{1\pm}\right]\exp\left(-i\Psi_{\pm}^{(0)}-\frac{\alpha(I)z}{2}\right), \tag{2}$$

$$E_{3\pm}(z)=\left[iJ_1\left(\Psi_{\pm}^{(1)}\right)E_{1\pm}-J_2\left(\Psi_{\pm}^{(1)}\right)E_{2\pm}\right]\exp\left(-i\Psi_{\pm}^{(0)}-\frac{\alpha(I)z}{2}\right), \tag{3}$$

$$E_{4\pm}(z)=\left[-iJ_1\left(\Psi_{\pm}^{(1)}\right)E_{2\pm}-J_2\left(\Psi_{\pm}^{(1)}\right)E_{1\pm}\right]\exp\left(-i\Psi_{\pm}^{(0)}-\frac{\alpha(I)z}{2}\right), \tag{4}$$

where $E_{1\pm}$ $(z)$ and $E_{2\pm}$ $(z)$ are the complex amplitudes of the circular components of the transmitted waves beams; $E_{3\pm}$ $(z)$ and $E_{4\pm}$ $(z)$ are the amplitudes of the self-diffracted waves, $E_{1\pm}$ and $E_{2\pm}$ are the amplitudes of the incident waves, $\alpha(I)$ is the irradiance dependent absorption coefficient, $I$ is the total irradiance of the incident beams, $J_m(\Psi_{\pm}^{(1)})$ stands for the Bessel function of order $m$, $z$ is the thickness of the nonlinear media, and

$$\Psi_{\pm}^{(0)}=\frac{4\pi^2 z}{n_0\lambda}\left[A\left(\left|E_{1\pm}\right|^2+\left|E_{2\pm}\right|^2\right)+(A+B)\left(\left|E_{1\mp}\right|^2+\left|E_{2\mp}\right|^2\right)\right], \tag{5}$$

$$\Psi_{\pm}^{(1)}=\frac{4\pi^2 z}{n_0\lambda}\left[AE_{1\pm}E_{2\pm}^*+(A+B)E_{1\mp}E_{2\mp}^*\right]. \tag{6}$$

We follow the notation $A=6\chi_{1122}^{(3)}$ and $B=6\chi_{1221}^{(3)}$, where the components of the third-order susceptibility tensor $\chi^{(3)}$ are related to the picosecond two-wave mixing as

$$\chi_{1111}^{(3)}=\chi_{1122}^{(3)}+\chi_{1212}^{(3)}+\chi_{1221}^{(3)}; \tag{7}$$

and the contribution to $\chi_{1111}^{(3)}$, given by the phase-mismatched single beam is described by [16]

$$\chi_{iiii}^{(3)}=\frac{1}{\varepsilon_o}\frac{\gamma_e}{\upsilon_a^2}\frac{\partial\varepsilon}{\partial\rho}=\frac{\varepsilon_o}{27\upsilon_a^2\rho}\left(n_o^2+2\right)^2\left(n_o^2-1\right)^2, \tag{8}$$

with

$$n_2=\frac{12\pi^2}{n_0^2 c}10^7\chi_{1111}^{(3)}, \tag{9}$$

where $n_2$ is the nonlinear refractive index in [cm$^2$/W], $\chi_{1111}^{(3)}$ in [esu], $n_o$ is the index of refraction at low intensity, $\varepsilon_o$ represents the permittivity of the vacuum, $\upsilon_a$ represents the acoustic speed through the media, $\rho$ represents the density of mass, $c$ represents the speed of light of the vacuum and $\gamma_e$ the electrostriction coefficient. Taking into account that in our

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experiments the irradiances of the self-diffracted waves are much lower than the irradiances of the incident waves, we used Eqs. (1-9) in order to identify the physical mechanisms that originate the nonlinearity of refractive index during the two-wave mixing, and then, we evaluate the contribution of the electrostriction force induced by a phase-mismatched beam.

## 3. Experiment

### *3.1 Processing route of the samples*

The silicon-nitride films were prepared using a remote plasma-enhanced chemical vapor deposition (RPECVD) system. The films were grown on quartz substrates using a working pressure of 300 mTorr, a substrate temperature of 300°C and a radio-frequency power of 200 W. The flow rates of $H_2$, Ar, $SiH_2Cl_2$ and $NH_3$ were 20, 150, 5 and 50 sccm (standard cubic centimeters per minute), respectively. During the deposition, Ar and $NH_3$ gases were fed from the top of the chamber where the plasma is formed; meanwhile, the $H_2$ and $SiH_2Cl_2$ gases were fed from the side and underneath the plasma by means of a dispersal ring located over the substrate holder. The silicon-nitride films were characterized by High-Resolution Transmission Electronic Microscopy (HRTEM).

### *3.2 Measurement of the different contributions to the nonlinear refractive index*

In order to measure the nonlinear optical response we used the experimental setup illustrated in Fig. 1, where L1-3 represent the focusing system of lenses, BS1-2 are beam splitters, λ/2 is a half-wave plate, M1-4 are mirrors, MNL is the sample, A1-2 are polarizers, and PD1-5 are photodetectors with integrated filters. The irradiance rate $I_1$:$I_2$ was 1:1 with a maximum pulse energy of 0.1mJ for each one, $\lambda$ = 532nm and 26ps with linear polarization. The radius of the beam waist at the focus in the sample was measured to be 0.15 mm. The beam $I_b$ was used for previous stimulation of the electrostriction effect associated with the samples, its maximum energy was of 0.15mJ. The mirrors M2 and M3 were located in a table with capabilities for micrometric motion, that together with the change in the position of BS1 and M1, it was possible to control the delay of the two-wave mixing interaction and the interaction of the beam $I_b$. The two-wave mixing was delayed until having the maximum magnitude of response without letting it interact with the other incident beam $I_b$.

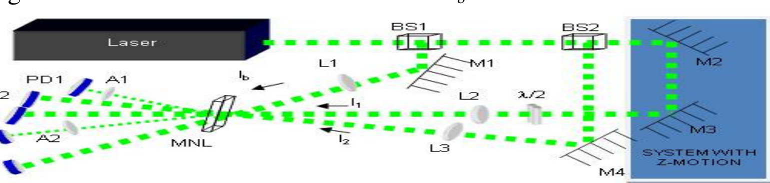


Fig. 1. Nonlinear optical experimental setup.

### *3.3 Estimation of the speed of sound by electrostriction measurements*

The change in the electrostriction effect that allowed us modulating the nonlinear refractive index was estimated by separated observations of self-diffraction originated by the two-wave mixing. Changes in the nonlinear refractive index can be obtained by a straightforward measurement of the self-diffracted signals and using Eqs. (1-9); considering the previous participation of the beam $I_b$ in the interactions into the sample.

## 4. Results and discussion

Figure 2(a) shows the typical HRTEM micrograph of the resulting thin film samples, where several dark spots that correspond to the Si-QDs can be observed. A statistical analysis of the HRTEM micrographs in different zones of the samples was made in order to get the size distribution of the Si-QDs. The results of this analysis showed that the Si-QDs average size is 3.2 nm and the correspondent Boltzmann fit is shown in Fig. 2(b). The optical transmittance spectrum is presented in Fig. 2(c). One can clearly observe an absorbing edge towards the UV

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that starts approximately at 290 nm, this is associated with the band gap of the sample containing the Si-QDs embedded in silicon-nitride and deposited in the quartz substrate.

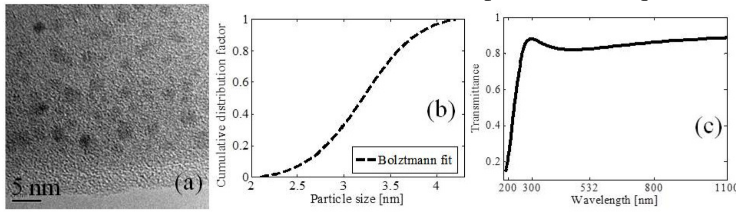


Fig. 2. (a) Typical HRTEM micrograph. (b) Statistical cumulative distribution of particle size. (c) Linear transmittance spectra.

In order to perform the nonlinear optical measurements, initially the beam $I_b$ was switched off in the experiments. Our measurement system was previously calibrated using carbon disulfide, $CS_2$, with a thickness of D = 1 mm, as a nonlinear medium with a well-known third-order nonlinear susceptibility of $\left|\chi^{(3)}\right| = 1.9 \times 10^{-12}$ esu [5]. In our case, the silicon-nitride sample has a thickness of D = 1.003 ± 0.12 µm. The polarized irradiances of the diffracted beams behind the sample were measured in two different cases of polarization of the incident beams; when the incident waves $\vec{E}_1$ and $\vec{E}_2$ had parallel and orthogonal linear polarizations. The axes of transmission of the analyzers A1-2 were aligned in order to detect, for each case, the parallel and the orthogonal components of the polarization of the waves. An error bar of ± 10% was estimated for the experimental irradiance data. By comparing numerical simulations of Eqs. (1-10) with the data obtained from the self-diffracted and transmitted irradiances in $CS_2$ and that obtained from the silicon-nitride samples we obtained an optical linear absorption of $\alpha_0 = 4.04 \times 10^5$ $m^{-1}$, a saturated absorption with $\beta = 7.7 \times 10^{-9}$m/W, an electronic nonlinear refractive index of $n_2 = 1.8 \times 10^{-16}$$m^2$/W, and $\left|\chi^{(3)}_{1111}\right| = 4.6 \times 10^{-10}$esu. Figure 3(a) shows the experimental (marks) and numerical (continuous lines) self-diffraction efficiency η, as a function of the angle $\phi$ between planes of polarization of the incident waves with the best fitting of the numerical simulations. These results are consistent with the reported values obtained in a similar sample with a different self-diffraction method [17]. Once it was identified the electronic response of the samples, we stimulated the electrostriction contribution to the nonlinearity of index by means of $I_b$. This phase-mismatched third beam produced a heightening in the self-diffracted signal, and for that reason, a higher contrast in the birefringence grating at the center of the interference pattern generated for the incident beams could be expected. Comparing the results for the pure electronic response of $n_2$ in our silicon-nitride samples, we measured an intensification of the self-diffraction efficiency of approximately 32% . The correspondent nonlinear refractive value given by the fitting of the experimental data are $\left|\chi^{(3)}_{1111}\right| = 1.822 \times 10^{-16}$ esu and $n_2 = 2.2 \times 10^{-20}$ $m^2$/W, for the electrostriction effect. Using Eqs. (8) and (9) with $n_o = 1.8$, and $\rho_o = 3180$ Kg/$m^3$ we estimated that the speed of the sound is $\upsilon_a = 3300$ m/s. The third order nonlinear response given by electrostriction appears six orders of magnitude lower than the electronic response. As a comparative result, we measured the nonlinear refraction for a pure silicon-nitride film and then we find that $\upsilon_a = 7800$ m/s. This last result is close to the acoustic speed measured for high frequencies in silicon-nitride from which, with a Young modulus (M) of 310GPa and with $\upsilon_a = (M/\rho_o)^{1/2}$, is equal to 9900 m/s [18]. A time-resolved wave-mixing technique was performed in order to maximize the resulting nonlinear refractive index associated with electrostriction. The maximum studied delay between the two-beam interaction and the interaction originated by the beam $I_b$ was about 2 ns; the best result was obtained for about 1.4 ns as it can be seen from Fig. 3(b). We calculate the change in the speed of sound in our sample by using Eqs. (8) and (9) and the numerical results based in

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experimental data are plotted as marks in Fig. 3(c) while the continuous line is a theoretical calculation; as it can be observed, a strong nonlinear modification on the speed of a mechanical wave can be achieved by an electrostrictive effect in a silicon-nitride film containing Si-QDs.

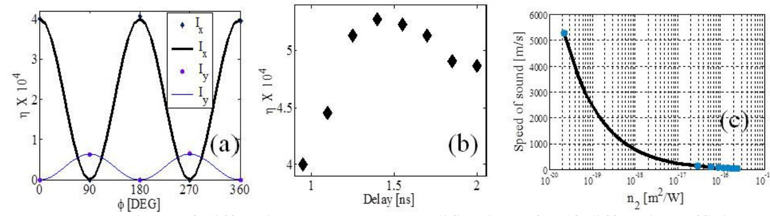


Fig. 3. (a) Vectorial self-diffraction results. (b) Modification of self-diffraction efficiency by the phase-mismatched beam. (c) Change in the speed of the induced mechanical waves vs. $n_2$.

It has been previously reported a nanosecond relaxation time of carriers from the indirect energy states in Si-QDs [19]; however a nanosecond Kerr homogeneous response in our sample was verified by a change in the linear polarization of the phase-mismatched beam through the experiments, it allows us to guarantee the absence of a birefringence that could be originated by a carrier phenomenon. Another argument that supports the disregarding of the responsibility of an electronic nanosecond nonlinear response in our experiments comes from the measurement and the evaluation of an ultrafast electronic nonlinear response in similar studied samples [17]. The enhancement of the nonlinearity of index in a silicon-nitride sample with the inclusion of Si-QDs has been previously associated to a quantum confinement effect [17]; however in this work, we notice that this circumstance of the inclusion of the Si-QDs also seems to produce an enhancement in the nonlinear electrostrictive response. This situation was experimentally supported when we compared the measurements in a pure silicon-nitride sample and no change in the self-diffracted signal was detected with the previous stimulation of a phase-mismatched beam.

Low dimensional semiconductor structures have originated numerous scientific researches in order to propose platforms for implementing diverse optoelectronic circuits, nevertheless the development of nanomechanical configurations for controlling high-speed signals also seems to be in a promising progress [2]. The optical technique described in this works presents the advantage to be a non-contact and non-invasive method for estimating or modulating the speed of sound in transparent dielectric media; besides, mechanical parameters like the Young Modulus or the density can be derived by inducing an electrostriction effect. Moreover it seems that these effects of nonlinearity of index can potentially exhibit functional applications in photothermal processes where it is mandatory to control the density for manipulating the propagation of induced mechanical waves.

## 5. Conclusion

We present a vectorial picosecond self-diffraction method for the optical evaluation of the speed of the sound in a silicon-nitride film containing Si-QDs prepared by RPCVD. Experimental results of the electronic and the electrostriction effects associated with the nonlinearity of index exhibited by the sample were obtained. Strong modifications in the propagation of acoustical waves were estimated and it seems that the nonlinear optical response related to the sample can be easily manipulated in order to control their mechanical response.


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