Experimental Observations of Bistability and Instability in a Two-Dimensional Nonlinear Optical Superlattice

Bin Xu^*

National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 2I 0008, People's Republic of China

Nai-Ben Ming

Center for Condensed Matter Physics and Radiation Physics, China Center of Advanced Science and Technology (World Laboratory), P.O. Box 8730, Beijing 100080, People's Republic of China and National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210008, People's Republic of China (Received 24 August 1993)

Optical bistability in a two-dimensional nonlinear superlattice, resulting from the index-modulation mechanism, was observed in experiment for the first time. The two-dimensional nonlinear superlattice was constructed by recording a two-dimensional refractive index grating into a photorefractive material, a Fe-doped LiNbO₃ single crystal. Optical instability was also observed in this experiment.

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Optical bistability in a one-dimensional (1D) superlattice containing Kerr-form dielectric nonlinearity was first proposed by the pioneering work of Winful, Marburger, and Garmire [1] more than ten years ago. This idea had then been developed in a number of theoretical papers [2-7]. Recently, Cada *et al.* [8] carried out an experiment using GaAs/A1As periodically layered structures and observed the all-optical logic operations. This may have been the first experimental attempt to demonstrate the bistable behavior in a 1D superlattice. The bistability in such a 1D periodic medium and that in a Fabry-Perot etalon are potentially analogous because of their dispersive natures in these two types of resonant structures, respectively. In a 1D superlattice, it is the change of the phase mismatch between the propagating wave vector and the periodic structure that brings the incident wave from a forbidden transmission state to an allowed state, or from a low-transmission state to a high-transmission state in the allowed band [4-7]. The bistability in a 1D superlattice is thus attributed to the phase-mismatch mechanism [9]. A recent theoretical work by the present authors has revealed that a novel bistable mechanism, i.e., the index-modulation mechanism, which is characteristically different from the phase-mismatch mechanism, exists in a two-dimensional (2D) superlattice containing Kerr-form dielectric nonlinearity [9]. It is known that in 1D superlattices the light's transmission in the exact phase-matched condition (the incident wave vector satisfies the exact Bragg condition) corresponds to the forbidden state [6,7]. However, in 2D superlattices, when the transmitting light satisfies the exact Bragg condition of multiwave diffraction, whether the transmission state is located in the forbidden band or in the allowed band, is determined by the values of a set of parameters which are defined as the index-modulation strengths of the 2D periodic structure [9,10]. If the values of these parame-

ters are arranged suitably, the light will propagate in the allowed band. Changes of these values will lead to highor low-transmission states for each diffracted wave, since in the allowed band in the exact Bragg diffraction condition, the intensities of the multidiffracted waves are oscillation functions of these parameters [9,10]. This element provides the basis of the index-modulation bistable mechanism, which is exhibited in 2D superlattices but not in 1D cases. When the Kerr-form dielectric nonlinearity is considered, as the theory in Ref. [9] predicts, the values of the index-modulation strengths will be perturbed by the interference of the transmission-diffraction light field in the medium, forming a positive feedback element to establish bistable behaviors in the incident-diffracted relations of the 2D superlattice [9].

To experimentally demonstrate this index-modulation mechanism, the photorefractive materials are ideal candidates. In these materials, the net refractive index change, caused by only the interference fringe of even a weak cw light field, has an expression similar to the Kerr-form dielectric nonlinearity. It will be potentially the same for the existence of the index-modulation bistable mechanism in photorefractive materials as in the Kerr-form nonlinear media [9]. The 2D superlattice in a photorefractive material can be constructed beforehand by recording a 2D refractive index grating into the medium with the volume holographic recording method. This is a convenient way to qualitatively, though not quantitatively, demonstrate the existence of the index-modulation mechanism in a 2D nonlinear superlattice. In this Letter, we report the first experimental observations of optical bistability, as well as instability, in such a 2D nonlinear superlattice.

The photorefractive material used in the experiment is an oxidized Fe-doped LiNbO₃ (0.1 wt. $%$ Fe) single domain crystal. A beam of the blue line (488 nm, TEM_{00} mode) of an argon-ion laser was split into three nearly-

FIG. 1. (a) Optical geometry for recording the 2D refractive index grating into a Fe-doped LiNbO₃ single domain crystal. (b) Schematic diagram of four-wave diffraction. I_0 is the incident wave satisfying the exact Bragg condition of four-wave diffraction.

equal-intensity ones, S_1 , S_2 , and S_3 , which were recombined and interfered to cause spatial variations of the refractive index. Thus a 2D square periodic grating was recorded into the crystal as Fig. 1(a) shows. (The indexmodulation strengths are Fourier components of the grating strength.) The intensities of the beams S_i ($i = 1,2,3$) were set to be about 2.5 W/cm^2 . The diameter of the beams, stipulating the size of the grating, was 0.2 cm. Despite the difficulty in determining precisely the grating strength in this holographic writing process, after an exposure time of about 30 to 40 s, the grating's diffraction efficiency reached a value in the range from 10% to 15% (the material's absorption is high). We should point out that sufficient high diffraction efficiency is a prerequisite for the following experiments. This oxidized Fe:LiNbO₃ sample showed high resistance to relax and the grating remained stable in the sample after the writing beams were moved. Then a beam I_0 , with the same wavelength and the same incident direction as the beam S_1 , was incident on this 2D grating with a much weaker intensity which was adjusted by an attenuator from 0 to 0.8 W/cm². Because this incident beam satisfied the exact Bragg condition, four diffracted waves were excited as Fig. 1(b) shows. This is the multiwave diffraction model the present authors used in the theoretical work [9] and here the allowed transmission is permitted. The photodiodes were used for measuring the diffracted intensities. With slowly adjusting the incident intensity I_0 and then recording the changes of the diffracted intensities with I_0 by the $X-Y$ recorder, the bistable behaviors would be clearly plotted if they appeared in the incident-diffracted relations.

In the illumination of the incident wave, the trapped electrons in the space charge pattern of the original grating are excited and drift. Because there is an interference field existing in the medium due to the incident wave diffracted into four directions, the space charge pattern that the drifting of the free carriers is constructing coincides with the original one. By the electric field induced

FIG. 2. With incident wave I_0 satisfying the Bragg condition of four-wave diffraction, and its intensity adjusted to form a cycle, the changings of the diffracted intensities I_2 and I_4 recorded. Intensity jumps occur simultaneously in the diffracted waves.

modulations on the refractive index, and because of the low illuminating intensity and the low erase sensitivity in this oxidized sample [11], this redistribution of the trapped electrons relative to the former actually gives perturbations to the values of the original indexmodulation strengths of the superlattice. The strengths of the perturbations depend on the incident intensity and they may be positive or negative, respectively, with different interference field. This fact provides the feedback element for the index-modulation mechanism which is expected to exist in such a 2D superlattice. The change of the values of the perturbed index-modulation strengths causes a change of the transmission-diffraction field, and this changed field will return to affect the perturbations of the index-modulation strengths. Diffracted intensities will thus change nonlinearly with the incident intensity. Figures 2 and 3 are two of the results recorded in experi-

FIG. 3. One result recorded in the same experimental process as that of Fig. 2, showing not only the intensity jump but also the behavior of instability. Inset: The intensity pulses periodically with time when I_0 is fixed in the region of instability. The pulsation period is of the order of 0.1 s, corresponding to the response time of the photorefractive effect in the sample.

FIG. 4. During the process of erasing the 2D grating by the illumination of the incident beam, discontinuous jumps of the diffracted intensities recorded in I_2 and I_4 . Notice that I_2 and 14 jump simultaneously and in opposite directions.

ments that demonstrate bistable behaviors with discontinuous jumps of the diffracted intensities. The hysteresis loops appearing here in the cycles of the figures are somewhat complicated to identify, for the redistribution of the trapped electrons in the space charge pattern is irreversible with the changing of the incident intensity and erasure occurs in this incident-diffracted process [12]. The experimental results may be improved by means of the fixing methods to fix the first-constructed 2D grating into the photorefractive sample [13,14]. In Fig. 3 it is noticed that the lower branch of the hysteresis loop develops into instability. As we analyzed above, the transmissiondiffraction field and the perturbed index-modulation strengths are interacting with each other. The observed bistable region in Fig. 2 indicates that this kind of interaction reached a stable state, and the stable state could be more than one; if this interaction did not reach a stable state, instability was exhibited as shown in Fig. 3,

To check the results further, in an experiment we improved the incident intensity and made it fixed at about 2 $W/cm²$. The sample's erase sensitivity increased a lot under such a high illuminating intensity. The 2D grating would be erased gradually and steadily with time when it diffracted the incident light. Then we recorded how the diffracted intensities were changed during this erasure process.

Figure 4 shows the experimental results observed in this erasure process. It is seen that the diffracted intensities are not always diminished continuously with time but with abrupt change or discontinuity, and behavior of instability was observed (see Fig. 5). This system can enter the regions of bistability or instability by means of either adjusting the incident intensity or changing the values of the index-modulation strengths.

The behaviors of instability result from the fact that the transmission-diffraction field in the medium and the perturbed index-modulation strengths cannot evolve to a convergent and self-consistent state with time. This happens when the parameters are in certain ranges. To verify that the index-modulation mechanism may lead to instability, here we borrow the model that we have used in the preceding theoretical work [9]. In the model, a self-

Erasing Time

FIG. 5. During the erasure process, instability observed in diffracted wave I_2 . Inset: Sine-wave periodic pulsations in the region of instability.

consistent solution of the iteration method is applied to achieve the stable state between the field and the perturbed index-modulation strengths; i.e., with numerical computation, the solution converges in a finite number of iterations. However, we find that when the incident intensity increases to certain values, the solution does not converge, but either pulses periodically [see Fig. 6(a)] or shows chaos [see Fig. 6(b)] with infinite numbers of iterations. Details about the instability in a 2D nonlinear superlattice can be investigated by solving a set of coupled partial differential equations that involve the field and the material's parameters, which we plan to discuss in a later paper.

In conclusion, we have observed experimentally for the first time the bistability and the instability of the indexmodulation mechanism in a 2D nonlinear superlattice that was constructed by recording a 2D refractive index grating into a photorefractive material, a Fe-doped Li- $NbO₃$ crystal. The experimental results here are just qualitative, and we find that the bistable hysteresis loops and the discontinuous jumps of the diffracted intensities are not as large as those of the theoretical calculations. These discrepancies may be due to the deviations of the experimental conditions from those in the theoretical model in these facts: (i) The writing and the incident beams are of nonuniform Gaussian profile, not the plane waves in the theoretical consideration. (ii) The nonlinear dielectricity in photorefractive materials is not exactly the same as that in the Kerr-form media. (iii) The volume holographic writing of a grating into a photorefractive material is in fact a dynamical process, the 2D grating deviated from an exact periodic one [13]. (iv) The material's absorption, effectively affecting the light transmission process, is not considered. We recommend that further experimental investigations to verify the

^I IG. 6. In the theoretical model of four-wave diffraction in a 2D square superlattice containing Kerr-form dielectric nonlinearity (see Ref. [9]) at constant incident intensity I_0 , the varying of the diffracted intensity I_2 obtained numerically in iterative processes. Divergent solutions appear instead of convergent self-consistent solutions between the perturbed index-modulation strengths and the transmission-diffraction field, such as (a) periodic pulsations at I_0 =2.23 and (b) chaos at I_0 =2.66. The setting of other parameters about the 2D superlattice follows exactly that in Ref. [9].

theory quantitatively may be carried out by using etching techniques, patterned epitaxy, etc., to fabricate 20 periodic structures in dielectric media with Kerr-form nonlinearity. This is also where the interest of such practical bistable devices lies.

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Present address: P.O. Box 2967l, Georgia Institute of Technology, Atlanta, GA 30332.

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- [14] If the grating is thermally fixed into the photorefractive sample, the electronic space charge pattern will be transformed into an ionic pattern that will not be affected by the light's illumination. If the incident power is strong and the sample's erase sensitivity is high enough, the perturbations of the index-modulation strengths of the electron originally brought by interference will change instantaneously with the changing of the field. Such reversible devices are practical for bistability. However, efforts should be made to achieve sufficient high diffraction efficiency after the thermal fixing.