

Chapter 2. Novel Processes and Materials for Infrared Nonlinear Optics

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2.1 Project Description

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Optical systems such as signal processors, limiters, spatial light modulators, and optical computers require large and fast optical nonlinearities. Although free carrier-induced nonlinearities in semiconductors have the picosecond speeds required for use in many applications, they were considered too weak to be practical for optical systems applications. In our work, we have demonstrated that these nonlinearities can be substantially enhanced in certain materials and in structures such as superlattices. We have recently observed the first optical nonlinearity caused by subband effects in InSb/AlInSb and HgTe/CdTe superlattices.

Superlattices have the potential for generating large optical nonlinearities because their band structures can be tailored to optimize particular nonlinear effects. Several novel nonlinear optical processes that utilize either the large subband nonparabolicity in the growth direction¹ or intersubband transitions in various configurations² specific to superlattices have been proposed. To our knowledge, these processes have not been observed.

With support from the National Science Foundation, we have observed optical nonlinearities caused by subband nonparabolicity in InSb/InAlSb and HgTe/HgCdTe superlattices by using four-wave mixing techniques with CO₂ lasers. We have observed an enhancement in the third-order susceptibility, $\chi^{(3)}$, when the laser light was polarized along the growth direction instead of in-plane. This experiment demonstrated that the superlattice subband structure induced larger optical nonlinearity than that of its bulk counterpart.³

The coupling between light and electrons is given by $\mathbf{p} \cdot \mathbf{E}$, where \mathbf{E} is the electric field of the light and \mathbf{p} the momentum of the n^{th} energy subband $\varepsilon_n(\vec{p})$. Therefore, to observe nonlinear optical effects caused by subband structures peculiar to superlattices, the electric field of the light must be polarized in the superlattice growth direction. We designated the direction perpendicular to the plane of the superlattice as \hat{z} and the in-plane direction as \hat{x} . Light polarized along \hat{x} probes only subband structures parallel to the superlattice plane, $\varepsilon_n(p_x)$, which is not different from their bulk counterparts. The interesting superlattice effects are contained in $\varepsilon_n(p_z)$; large enhancement of p_z -nonparabolicity has been predicted. Furthermore, intersubband transitions are induced by E_z but not E_x .

¹ W.L. Bloss and L. Friedman, *Appl. Phys. Lett.* 41:1023 (1982); S.Y. Yuen, *Appl. Phys. Lett.* 43:813 (1983); G. Cooperman, L. Friedman, and W.L. Bloss, *Appl. Phys. Lett.* 44:977 (1984); Y. Chang, *J. Appl. Phys.* 58:499 (1985).

² S.Y. Yuen, *Appl. Phys. Lett.* 43:813 (1983); D.J. Newson and A. Kurobe, *Appl. Phys. Lett.* 51:1670 (1987).

³ D. Walrod, S.Y. Auyang, and P.A. Wolff, *Appl. Phys. Lett.* 56:218 (1989).

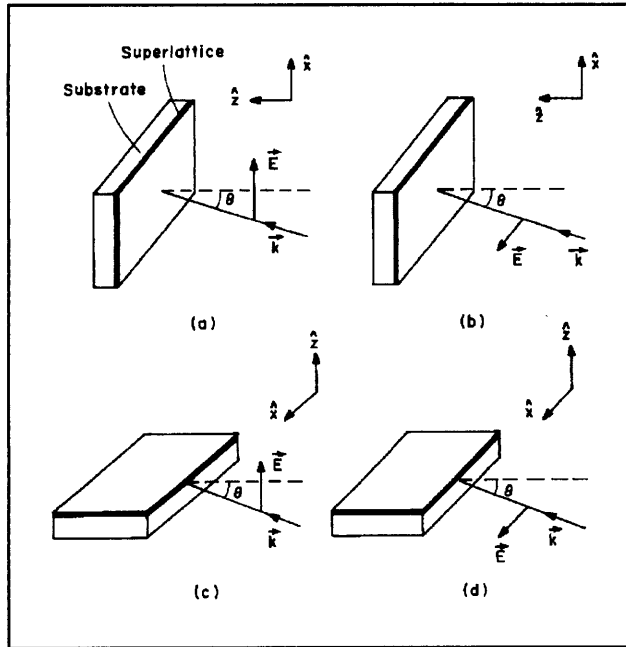


Figure 1.

The coupling to $\epsilon_n(p_z)$ is difficult to achieve in conventional nonlinear optical experiments in which light impinges upon the surface of the sample (figure 1). Because of the large indices of refraction of semiconductors E_z inside the sample, there was never more than a small fraction of the total field. Even at Brewster angle incidence, less than ten percent of the light intensity was polarized along \hat{z} since the nonlinear signal is proportional to the cube of the input intensity. Therefore, the third-order nonlinear susceptibility $\chi^{(3)}$ due to $\epsilon_n(p_z)$ must be at least fifty times larger than the bulk susceptibility to be distinguished in this geometry. We predicted these large susceptibilities for intersubband transitions when the subband gap was exactly matched to the photon energy.⁴ Intersubband transition was a resonant effect, however, and the predicted $\epsilon^{(3)}$ might not be achievable if the actual subband separation failed to match the photon energy. Optical nonlinearities generated by the non-

parabolicity of the lowest subband $\epsilon_0(p_z)$, were nonresonant, not placing as severe a demand on the specification of the sample. However, the enhancement over the bulk was smaller.⁵

We observed optical nonlinearities caused by subband structures in the growth direction in an AlInSb/InSb superlattice by using an end-firing technique. The AlInSb/InSb superlattice we studied consisted of 50 periods, each with 45 Å of InSb and 60 Å of Al_{0.08}In_{0.92}Sb. This superlattice was separated from the 350 μm Cr-doped GaAs substrate by a 200 Å AlSb buffer layer. The samples were undoped. We used a 50 μm InSb epilayer grown under the same conditions on the same kind of substrate for a control sample.

For the end-firing experiments, we cleaved off a strip about 1 mm in width. The cleaved edge provided good transmission. We observed about 50 percent transmission through the 1-mm sample; almost all loss was due to reflection. To ensure that the GaAs substrate did not contribute to the nonlinear optical signal, we cut off small sections of the samples and polished off the superlattice. Then we compared the substrate with a sample in which the superlattice was left intact. In all cases, the nonlinear signal vanished when we removed the superlattice.

The end-firing configuration enabled us to probe the \hat{z} -direction dispersion relation by changing the polarization direction. The four-wave signal from the InSb epilayer does not change as the light polarization is rotated 90 degrees. This result suggests that the material is isotropic. The four-wave signal from the superlattice was comparable to that of the InSb epilayer when the light was polarized along \hat{x} . However, as the polarization was rotated to the \hat{z} direction, the four-wave signal increases by twenty-two times, as depicted in figure 2.

⁴ S.Y. Yuen, *Appl. Phys. Lett.* 43:813 (1983); D.J. Newson and A. Kurobe, *Appl. Phys. Lett.* 51:1670 (1987).

⁵ W.L. Bloss and L. Friedman, *Appl. Phys. Lett.* 41:1023 (1982); S.Y. Yuen, *Appl. Phys. Lett.* 43:813 (1983); G. Cooperman, L. Friedman, and W.L. Bloss, *Appl. Phys. Lett.* 44:977 (1984); Y. Chang, *J. Appl. Phys.* 58:499 (1985).

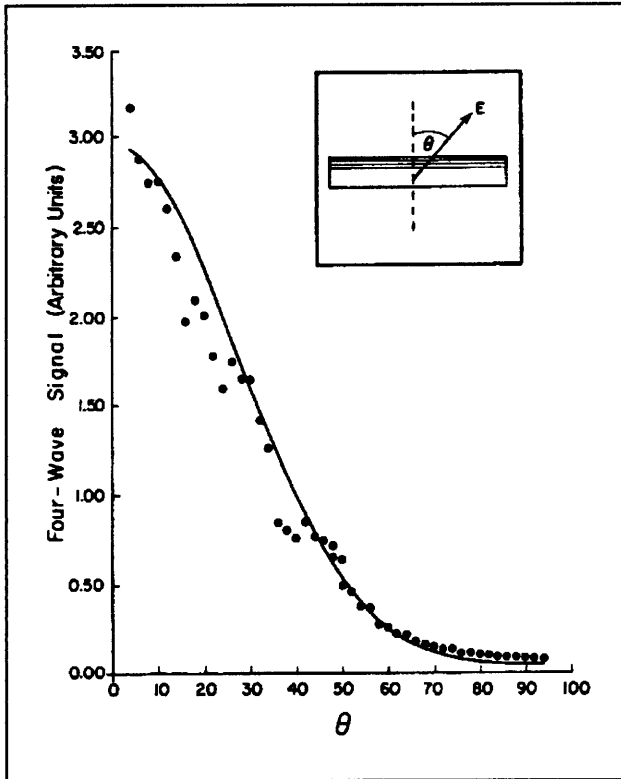


Figure 2.

By comparing the optical nonlinearities generated by different light polarizations, we ascertained that the \hat{z} -direction subband structure gave an enhancement of $\chi^{(3)}$ over the bulk value. There are several possible explanations for the enhancement, but we were unable to distinguish them quantitatively. Our research is continuing in this direction.

Publications

Auyang, S.Y., and P.A. Wolff. "Free-Carrier-Induced Third Order Optical Nonlinearities in Semiconductors." *J. Opt. Soc. Am. A* 6:2696 (1989).

Walrod, D., S.Y. Yuen, P.A. Wolff, and W. Tsang. "Optical Nonlinearities due to Subband Structures in $\text{Al}_{0.08}\text{In}_{0.92}\text{Sb}/\text{InSb}$ Superlattices." *Appl. Phys. Lett.* 56 (3):218-220 (1989).

