Quadratic optical nonlinearity of an amorphous heterojunction

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It has been shown experimentally, in the particular case of second-harmonic generation in a multilayer planar optical waveguide, that an amorphous heterojunction can have a significant quadratic nonlinearity. The possibility of "constructing" nonlinear media from ultrathin amorphous films is demonstrated.

A medium having a symmetry center has no nonvanishing components in its dipole quadratic nonlinear-susceptibility tensor, and the second-order nonlinear processes observed in such a medium stem from the leading terms in the expansion of the nonlinear-polarization vector.[1] In the region of disrupted local symmetry near the surface of a medium which is centrally symmetric in its interior, the components χ^{s}_{ijk} of the dipole quadratic nonlinear-susceptibility tensor are determined by a sum of "field" and "structural" components.[2,3]

$$\chi^{s}_{ijk} = \chi^{(3)D}_{ijkl} E^{0}_{l} + \frac{1}{2} \nabla_{l} \chi^{(2)Q}_{ijkl}, \tag{1}$$

where $\chi^{(3)D}_{ijkl}$ and $\chi^{(2)Q}_{ijkl}$ are the components of the third-order dipole nonlinear-susceptibility tensor and the second-order quadrupole nonlinear-susceptibility tensor. The first term in (1) stems from the presence of an electrostatic field $\vec{E}^{0}$, while the second stems from the spatial inhomogeneity of the medium near its surface.

The field component can play an important role in nonlinear processes near an interface between amorphous materials.

During the fabrication of an amorphous heterojunction, the junction may be subjected to external agents which cause an intense generation of electron–hole pairs. Mobile carriers would be injected, and a space–charge region would form. After the external agent stops acting, the charges would be "frozen" for a long time in localized states near mobility gaps of the amorphous materials. At a high density of the nonequilibrium charge in trapping levels, the electric fields in the space–charge region may exceed[4] 10^{7} V/m. From the typical values of the third-order nonlinear susceptibility for condensed media, 10^{-22}–10^{-21} m^{2}/V^{2}, and an internal field of 10^{8} V/m, we can find an estimate of 10^{-14}–10^{-13} m/V for the quadratic nonlinearity.

Sotin *et al.*[5] have detected second-harmonic generation in a multilayer amorphous structure consisting of two periodically alternating ultrathin insulating films. Such a structure could be interpreted as a medium with a high density of atomically sharp[6] heterojunctions "connected in opposition." The symmetry of the boundary region in each layer, bracketed by essentially identical media on the two sides, is locally lowered

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to a $C_{\infty v}$ macroscopic symmetry. The polar axes in the regions near the upper and lower boundaries of the layer are in opposite directions, while the absolute values of the components of the tensor $\vec{\chi}^s$ are nearly identical.

Our purpose in the present study was to reduce the degree of mutual cancellation of the contributions of the upper and lower boundary regions of each layer to the second harmonic. This reduction would make it possible to substantially increase the nonlinearity of the amorphous heterostructure as a whole and to find an estimate, possibly on the low side, of the quadratic nonlinear susceptibility of the amorphous heterojunction. It was proposed for this purpose that the period of the structure be formed from three amorphous films.

The structures were fabricated by reactive rf cathode sputtering. Targets of tantalum, aluminum, and fused quartz were sputtered in an oxygen–argon atmosphere. The substrates were moved in succession into the regions in which the various targets were sputtered. The resulting samples were amorphous according to x-ray measurements.

As an optical layout for achieving second-harmonic generation, we selected a planar waveguide which made it possible to achieve a high power density of the pump wave over a substantial interaction distance. The optical anisotropy of a multilayer periodic structure made of materials differing in refractive index[7] made it possible to match the fundamental waveguide modes, TE_0^{ω} and $TM_0^{2\omega}$ (Refs. 5 and 8).

The second-harmonic generation was observed in a waveguide structure 0.70 μm thick on a fused quartz substrate. Each of the 15 periods consisted of a Ta_2O_5 film 19.0 nm thick, an Al_2O_3 film 12.0 nm thick, and an SiO_2 film 15.7 nm thick. The source of the pump light, with a wavelength of 1.06 μm, was a free-running Nd:YAG laser (Fig. 1). The pump light and the second harmonic were coupled out of the waveguide completely by means of prism *7*, which was placed at fixed positions, at steps of 1 mm, along the X axis.

The results of these measurements are shown in Fig. 2. At the point at which the ratio $P^{2\omega}/(P^{\omega})^2$ reaches its maximum, the efficiency of the second-harmonic genera-

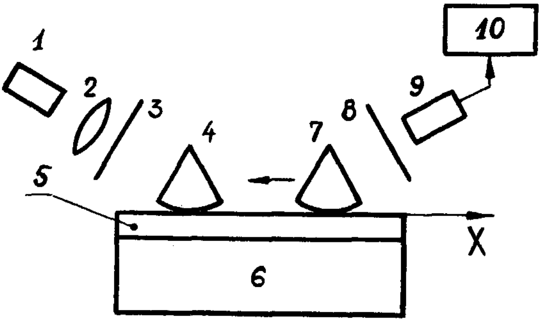

FIG. 1. Layout of the experiment on second-harmonic generation. *1*–LTI P Ch-4 laser; *2*—lens with $f = 0.5$ m; *3*, 8—filters; *4*, *7*—prism input and output coupling elements (TF-5); *5*—amorphous heterostructure; 6—substrate; *9*—FÉU-51 photomultiplier; *10*—S1-79 oscilloscope. The arrow shows the direction in which the output coupling prism is moved.

tion, $\eta = P^{2w}/P^{w}$, is 1.7×10^{-6} at a pump-wave power $P^{w} = 130$ mW.

Measurements of the longitudinal components of the wave vectors of the pump wave, k_x^{w} and the harmonic, k_x^{2w}, showed that the waveguide is nonuniform in the second-harmonic generation region and that the dependence $[k_x^{2w}(x) - 2k_x^{w}(x)]/k_0^{2w}$, which characterizes the phase detuning $(k_0^{2w} = 2w/c)$, is approximately linear, with $\Delta(\delta_\gamma)/\Delta x \approx 1.0\times10^{-3}$ cm^{-1}.

Using the approximation of a uniform distribution of the component d_{31} (we are assuming that the Z axis runs perpendicular to the plane of the substrate) in a cross section of the periodic structure, we calculated an average value over a period: $d_{31}^{\mathrm{av}} = 0.33\times10^{-13}$ m/V.

We also fabricated a control structure, with a "symmetric" bracketing of layers, under the same conditions. The twelve periods of this control structure consisted of films of Ta_2O_5 (19.0 nm) and SiO_2 (24.3 nm). Again in this control structure we observed a second-harmonic generation. The conversion efficiency, corrected to a pump-wave power of 1 W, was lower by four orders of magnitude than that found in the "asymmetric" structure (Fig. 2). The value of d_{31}^{av} was lower by a factor of 30.

In summary, these experiments have demonstrated that an amorphous heterojunction can exhibit a significant quadratic nonlinearity. The method of second-harmonic generation,[2] combined with methods for measuring the internal field,[4] may prove to be a useful tool for obtaining important information about the structural defects in the space-charge region of an amorphous heterojunction.

An increase in the degree of asymmetry of the distribution of the component d_{31}

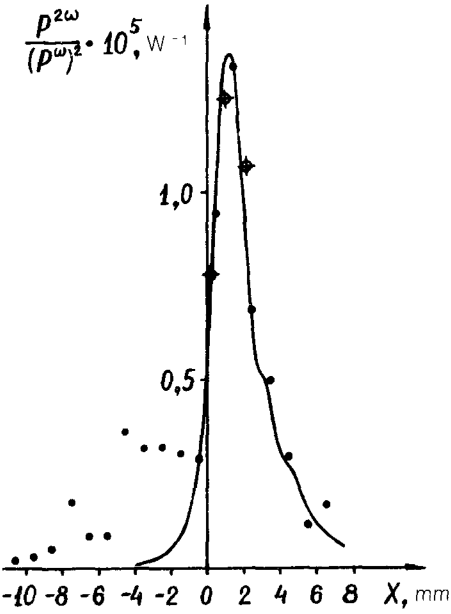

FIG. 2. Distribution of the efficiency of the second-harmonic generation, corrected to a 1-W power of the pump wave. Solid line—Calculated; filled circles—experimental results on the "asymmetric" structure; circles with plus signs—the same, for a "symmetric" structure($\times10^4$). The calculation was carried out under the assumption $\Delta(\delta\gamma - /\Delta x = 1.0\times10^{-3}$ cm^{-1} with $\delta\gamma(0) = 0$. The waveguide loss of the second harmonic was 21 dB/cm. The effective length of the radiation aperture was 1.5 mm.

in the cross section of each individual layer has resulted in an increase by a factor of 30 in the average value of this component over the period of a multilayer amorphous heterostructure. In the same manner it has been demonstrated that it is possible to "construct" nonlinear media from ultrathin amorphous films. With a substantially greater variety of materials to choose from in the fabrication of (for example) nonlinear integrated-optics devices, the principle proposed here would also make it possible to match the low-index waveguide modes of the pump and the second harmonic and to reverse the polar axis of a multilayer structure by varying the order of the constituent layers during the fabrication process.

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