Photoluminescence of quantum-size-effect epitaxial films and structures of $Pb_{1-x}Sn_xTe$

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A quantum size effect has been discovered in the photoluminescence spectra of thin films and multilayer heterostructures of $Pb_{1-x}Sn_xTe$. The spectrum, which consists of a large number of lines, is shifted in the high-energy direction by elastic strain in the samples.

The first $Pb_{1-x}Sn_xTe$ ($0 \leqslant x \leqslant 0.2$) superlattices were produced by laser deposition[1] and the "hot-wall" method[2,3] and used to study the transmission spectra and the magnetoresistance. A laser operating by virtue of quantum-size-effect PbTe films has been fabricated by molecular beam epitaxy.[4]

In the present letter we report the first observation of a photoluminescence of quantum-size-effect epitaxial films of *p*-PbTe and of multilayer heterostructures *p*-PbTe/*p*-$Pb_{0.89}Sn_{0.11}Te$ grown on (111) BaF_2 substrates. Thin PbTe layers (200–3000 Å) are synthesized by electrolytic etching of thick layers ($L \cong 2\ \mu$m) or are grown by the hot-wall method. The multilayer heterostructures are grown by instantaneous evaporation in a vacuum. They consist of a PbTe buffer layer with a thickness of 0.15–0.3 μm and have five or 20 pairs of alternating layers of PbTe and $Pb_{0.89}Sn_{0.11}Te$ of identical thickness ($\sim$400 Å). A YAG laser is used for excitation.

The photoluminescence is observed both in thin layers and in multilayer heterostructures at relatively high excitation levels ($\gtrsim 10^5$ W/cm^2). The emission is a stimulated emission. Its intensity is comparable in order of magnitude to that of the photoluminescence of thin films of PbTe and $Pb_{0.89}Sn_{0.11}Te$ at the same excitation levels. The photoluminescence spectra of both thin layer (Fig. 1) and multilayer heterostructures (Fig. 2) reveal large shifts (up to 20 meV) of the long-wave emission edge toward higher energies. With increasing excitation level, the width of the spectra increases reaching 60 meV. In these spectra we observe a fine structure in the form of single lines.

The large shifts in the photoluminescence spectra are attributed to an elastic strain in samples of this type. A quantization of the spectrum also leads to an increase in the width of the band gap, E_g, but these shifts are far smaller (1–8 meV). A strain arises for two reasons: the difference (Δa) in the lattice constants and a difference in the coefficients of linear thermal expansion (α) for BaF_2, PbTe, and $Pb_{0.89}Sn_{0.11}Te$. The large ($\Delta a/a \sim 4\%$) mismatch of the PbTe and BaF_2 lattice constants is erased in the course of the growth primarily because of the formation of mismatch dislocations at the interface between the thin layer and the substrate. In the case of the multilayer heterostructures, the mismatch dislocations remain for the most part in the buffer layer. The slight mismatch ($\Delta a/a \sim 0.23\%$) of the lattice constants in the adjacent

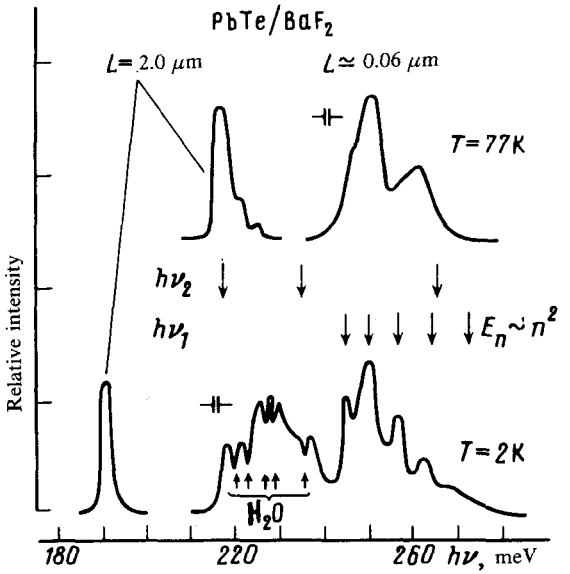

FIG. 1. Photoluminescence spectra of a thin ($L \simeq 600$ Å) layer of PbTe/BaF_2 at 2 and 77 K. Shown for comparison at the left are the photoluminescence spectra of a thick ($L = 2\,\mu$m) layer at the same temperatures. The arrows show H_2O absorption lines.

semiconducting layers is erased by the elastic extension of the $Pb_{0.89}Sn_{0.11}Te$ layers and by the elastic compression of the PbTe layers.

Since $\alpha(BaF_2) < \alpha(PbTe)$, the thin layers of PbTe on BaF_2 become stretched as the samples are cooled to low temperatures. An initial compression in the PbTe layers in the multilayer heterostructures is offset by the stretching during cooling, while in the $Pb_{0.89}Sn_{0.11}Te$ layers the stretching is even more pronounced. Diffractometric measurements in the superlattices at a low temperature (25 K) have shown[3] that the

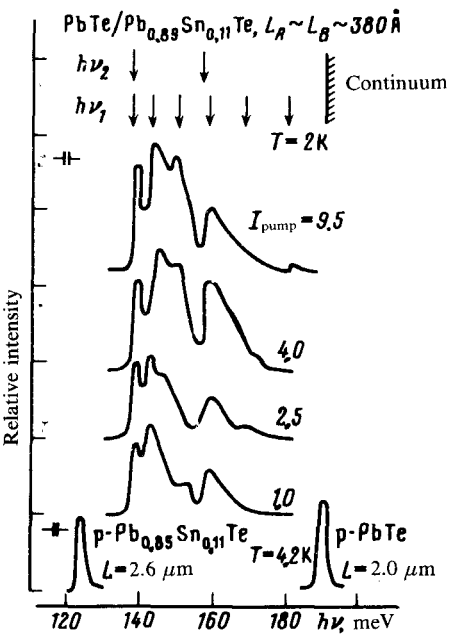

FIG. 2. Photoluminescence spectra of a PbTe/$Pb_{0.89}Sn_{0.11}Te$ multilayer heterostructure at 2 K as a function of the excitation level. Here L_A and L_B are the layer thicknesses. Shown for comparison at the bottom are photoluminescence spectra of thick ($L \geqslant 2\,\mu$m) PbTe and $Pb_{0.89}Sn_{0.11}Te$ layers at 4.2 K.

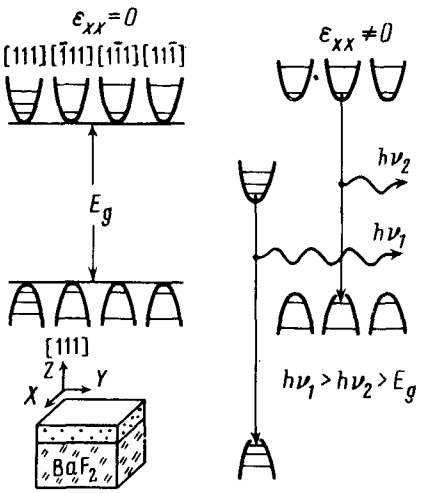

FIG. 3. Effect of a uniaxial strain on the $Pb_{1-x}Sn_xTe$ ($0 \leqslant x \leqslant 0.2$) energy spectrum. The inset shows the geometry of the sample.

PbTe layers are essentially unstressed, while the PbSnTe layers are under a tensile stress. At a low temperature, the thin layers of PbTe on BaF_2 and the layers of $Pb_{0.89}Sn_{0.11}Te$ in the multilayer heterostructure are thus stretched in the plane of the layers.

This stretching is equivalent to a uniaxial strain along the [111] direction. The uniaxial strain lifts the intervalley degeneracy, and the four equivalent L valleys split (Fig. 3) into a [111] main valley and three inclined valleys. [$\bar{1}$11], [1$\bar{1}$1], and [11$\bar{1}$]. Working from the shifts in the photoluminescence spectra and the strain energies,[3] we find a strain $\epsilon_{xx} = 3.4 \times 10^{-2}$ for a thin PbTe/BaF_2 layer and a strain $\epsilon_{xx} = 5.5 \times 10^{-3}$ for a $Pb_{0.89}Sn_{0.11}Te$ layer in a multilayer heterostructure. In the latter case, the stress is partially removed by the buffer layer. In a thin layer of PbTe/BaF_2, the value of E_g increases from 189 to 245 meV in the main valley and to 211 meV in the inclined valleys.

Because of the small thickness of the layers, these samples are quantum-size-effect entities. The thin PbTe/BaF_2 layer serves as a good model of a square potential well with infinite walls. The multilayer heterostructures constitute a set of isolated potential wells because of the rather large thickness of the barriers ($\sim$400 Å). In the effective-mass approximation, the positions of the energy levels in the quantum wells with respect to the band extremum are $E_n = \hbar^2 k_z^2/2m^*$, where the wave vector along the quantization direction, k_z, can take on only the discrete values $n\pi/L$, where n is the index of the level. Since the effective mass along the [111] quantization direction in the main valley is larger than in the inclined valleys, there are two sets of energy levels, with different level spacings, in each potential well. We thus find two groups of possible optical transitions, shifted with respect to each other by the uniaxial strain. In the photoluminescence spectrum of the thin PbTe/BaF_2 layer (Fig. 1; $T = 2$ K), there are in fact two groups of lines; the group at the right is described well by $E_n \sim n^2$ with the selection rule $\Delta n = 0$. The energy positions of the photoluminescence lines agree with the results of a calculation from a two-band model.

To determine the energy of the optical transitions in the multilayer heterostruc-

ture, we need to take into account the finite height of the barriers, which furthermore changes upon the imposition of a strain. To calculate the barrier height, we use the $Pb_{1-x}Sn_xTe$ energy diagram from Ref. 3. As in the thin PbTe/BaF_2 layers, we find two sets of optical transitions, which overlap to a greater extent because of the lesser strain of the layers. The results of these calculations also agree satisfactorily with the positions of the photoluminescence lines in the spectra (Fig. 2).

The energy positions of the photoluminescence lines and the approximately equal intensities of the emission from the main and inclined valleys are evidence that the hot ($h\nu_{laser} \gg E_g$) nonequilibrium current carriers relax to both the main and inclined valleys. Since this a simulated photoluminescence, with a scale time $\tau \sim 10^{-10}$–10^{-11} s, the carriers do not have time to become redistributed through intervalley scattering, and a common Fermi quasilevel is not established in the main and inclined valleys. For this reason, we observe two groups of optical transitions in the photoluminescence spectra.

The integral intensity of the photoluminescence of the multilayer heterostructures is comparable to the photoluminescence intensity of thick PbTe and $Pb_{0.89}Sn_{0.11}Te$ films at identical excitation levels. This result is evidence in favor of the contravariant diagram of the PbTe/PbSnTe heterojunction proposed in Refs. 3 and 5, while it contradicts data[6] on the composition dependence in $Pb_{1-x}Sn_xTe$ of the positions of the L_6^+ and L_6^- terms, which form a band gap.

In summary, these experiments have revealed, for the first time, a photoluminescence of thin, quantum-size-effect layers of PbTe and of multilayer heterostructures of the PbTe/$Pb_{0.89}Sn_{0.11}Te$ type. The existence of a fairly intense photoluminescence of these heterostructures is evidence in favor of a contravariant diagram of the PbTe/$Pb_{0.89}Sn_{0.11}Te$ heterojunction. Since thin layers of PbTe and multilayer heterostructures grown on BaF_2 substrates are stressed, the photoluminescence spectra are shifted significantly up the energy scale.

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