Journal of Alloys and Compounds, 195 (1993) 187–190
JALCOM 7444

GROWTH AND OPTICAL STUDY OF SUPERCONDUCTING SUPERLATTICES

I.E. Trofimov[*] H.-U.Habermeier, A.P.Litvinchuk[†] D.H.Leach, K.Kamarás[‡] C.Thomsen, and M.Cardona
Max-Planck-Institut für Festkörperforschung,Stuttgart 80 ,Germany.

Abstract.

Raman scattering and far-infrared reflectivity experiments have been performed on the YBCO-based superconducting superlattices. The data obtained indicate on the reconstruction of the electronic system in the superlattices and on its dependence on SL parameters.

1 Introduction.

Recently, the epitaxial growth of high temperature superconducting superlattices (SL), especially YBCO/-PrBCO has been obtained [1,2]. Superlattice research has been focused on interlayer coupling and superconducting properties of a single unit cell. It has been found that SL with an individual thickness of YBCO layers only one unit cell along the c-axis remain superconducting with $T_c \sim 40K$. There is now a large amount of interesting data on the transport properties of these multilayer structures. However, the reduction of T_c, has given rise to different explanations. Venkatesan et al.[3] proposed that the decrease of T_c is related to the decoupling of the YBCO layers, while Wood [4] gave an alternative explanation based on a "hole filling" model. These two models do not exhaust all the possible reasons for lowering T_c. For example, the transformation of phonon spectra in SL might be responsible for this effect if a phonon coupling mechanism takes place. In addition, the important question concerning the relationship between the superconducting energy gap (Δ) and T_c in SL is still open. Optical methods are traditionally used not only for studying the superconducting energy gap directly, but also to elucidate the nature of the excitations which mediate the pairing [5,6]. As far as high-temperature superconductors (HTSC) are concerned, the various procedures proposed give contradictory results and there is still no agreement on how to extract the value of Δ from infrared and Raman measurements [5,6].

2 Sample preparation and characterization.

YBCO-based SL with barrier layers of PrBCO and $Pr_xY_{1-x}BCO$ were made with a pulsed-laser deposition technique, which is described elsewhere [7]. Stochiometric targets were mounted on a multitarget holder that permits individual rotation of the targets as well as their exchange during the deposition process. The SL were grown on $SrTiO_3$, $NdGaO_3$ and $LnAlO_3$ (100) substrates at a deposition rate of 0.5 - 1.5 Å/pulse and 2 Hz in an ambient O_2 pressure of 0.2 - 1 mbar, with the substrate temperature in the range of 670 - 800 C. A pulsed KrF (λ=248 nm) eximer laser produced an energy density in the targets of about 1.7 J/cm^2. The substrate-target distance was kept at 4 cm. The substrate rotation during deposition has significantly improved homogeneity and thickness uniformity of the samples. After the growth procedure the SL were cooled within 30 minutes to room temperature at an oxygen pressure of 1 bar. Resistivity measurements reveal the well-known decrease of T_c and the broadening of the transition with decreasing of the YBCO layer thickness [3]. The structure of the SL was examined by means of X-ray diffraction. Fig.1 shows a diffraction pattern of a SL YBCO(7ML)/PrBCO(7ML). Only (*ool*) diffraction peaks are observed indicating complete c-axis texturing. The satellite peaks on both sides of the main ones are due to an additional periodicity in the SL structure. The position of satellite peaks is related to the SL period as follows:

$$\Lambda = (\lambda/2)[sin\Theta_i - sin\Theta_{i-1}]^{-1},$$

---

[*] **Permanent address:P.N.Lebedev Physics Institute, 117924 Moscow, Russia**
[†] **Permanent address: Institute of Semiconductors, 252650 Kiev-28, Ukrain**
[‡] **Permanent address: Research Institute for Solid State Physics, H-1525 Budapest 114, POB 49, Hungary**

0925-8388/93/$06.00

where λ is the x-ray wavelength (1.542 Å in our case) and the Θ_i are the positions of two adjacent satellite peaks. The calculated values correspond well to that derived from direct measurements of the SL thickness. From measurements of the Raman intensities it is possible to determin the degree of epitaxy (η) [8]. We have found $\eta \geq 0.88 \pm 0.02$ in the case of c-axis oriented SL.

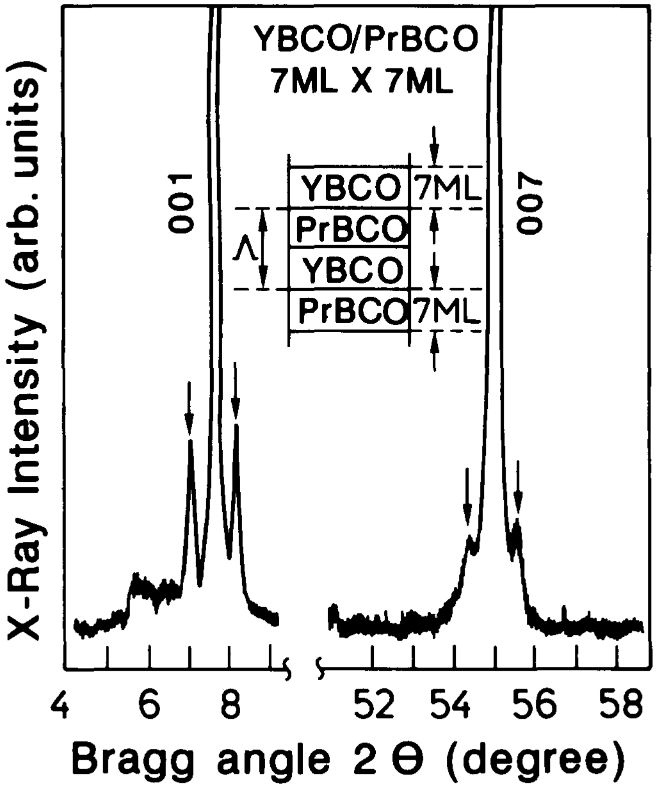

Figure 1: X-ray diffraction pattern for the SL YBCO(7ML)/PrBCO(7ML)

3 Raman.

In Fig.2 spectra of c-oriented YBCO, PrBCO, $Y_{0.5}Pr_{0.5}BCO$ and SL YBCO/PrBCO are shown. One can see that unlike the alloy, there are two sharp B_{1g}-like phonon peaks at 304 and 344 cm^{-1} in the spectrum of a SL. All the peaks in SL spectra are as narrow as in the bulk materials, indicating good crystallinity and abrupt interfaces without any detectable disordering due to interdiffusion or any other technological imperfectness. An important feature of the phonon spectra is the softenning below T_c of the B_{1g} mode confined in YBCO layers (Fig.3), which is a typical property of bulk superconductors [6]. However, the shape of the B_{1g} YBCO line in the SL is more symmetric than in the bulk. The intensity of the background in the SL case is also approximatly two times lower than in crystals (compared to the phonon intensity).The phonon asymmetry, one believes, is a result of the interaction of phonons with a broad electronic continuum having the same symmetry (Fano effect).

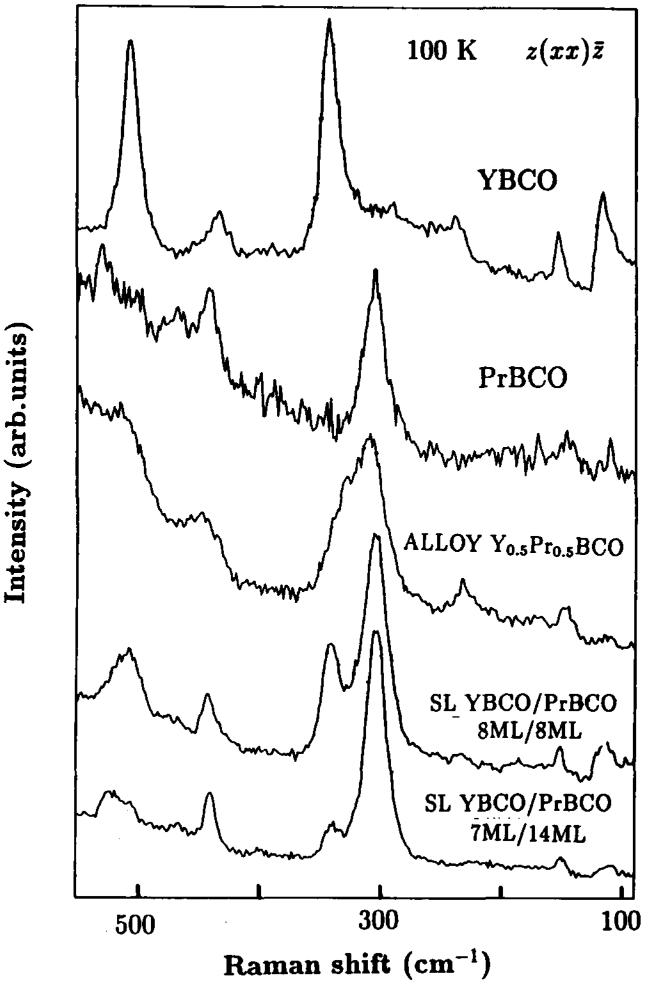

Figure 2: Raman spectra of the bulk YBCO, PrBCO, $Y_{0.5}Pr_{0.5}BCO$ alloy and YBCO(8ML)/PrBCO(8ML), YBCO(7ML)/PrBCO(14ML) superlattices taken at 100 K in the $z(xx)\bar{z}$ geometry.

The same qualitative changes in the spectra with respect to bulk YBCO were found for all SL. As far as the absolute value of the softening is concerned, it depends strongly upon the YBCO layer thickness and diminishes continuously from about 8 cm^{-1} for the thick YBCO film to 1 cm^{-1} for the SL 3ML/3ML. Phonons confined in the PrBCO layers show, however, no softening. The vanishing of the Fano effect in SL may indicate a decrease of the free electron contribution to the continuum excitations via charge redistribution between layers or localization.

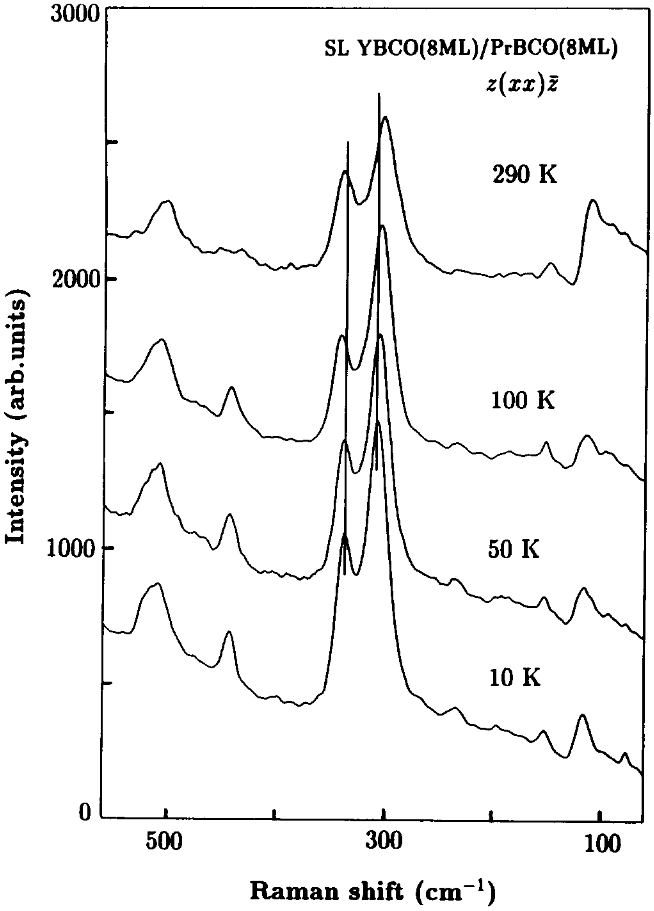

Figure 3: Raman spectra of the SL YBCO(8ML)/PrBCO)(8ML) in the $z(xx)\bar{z}$ geometry at room temperature, 100, 50 and 10 K. The curves are shifted by 500 on the vertical scale. Solid lines mark the low-temperature position of the B_{1g}-modes confined YBCO- and PrBCO-layers .

The direct scattering of light by electronic excitations gives important information about a superconducting gap. In HTSC, gap formation is usually related with the suppression of a low-energy scattering intensity at temperatures below T$_c$ [9]. The roll-off point, at which the intensity decrease begins, one takes for the value of Δ. Low-temperature spectra o the SL YBCO(3ML)/ PrBCO(3ML) are illustrated in Figs.4 and 5. According to Raman scattering data, the coupling constant $2\Delta/k_BT_c$ in YBCO is about 4-5, hence we would expect a "gap" peculiarity in the spectra of this SL near 200 cm^{-1}. However, the SL does not show any suppression of the scattering intensity in both B_{1g} and A_{1g} symmetries down to 100 cm^{-1}. The measured behavior argues against an assignment of the roll-off point in the spectra to the gap feature.

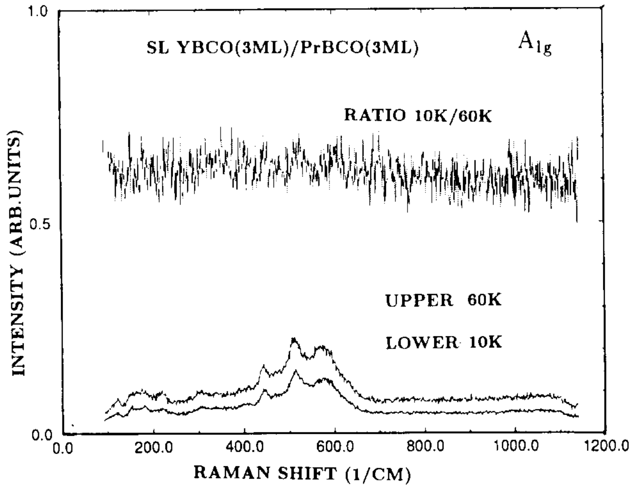

Figure 4: Raman spectra of the SL YBCO(3ML)/ PrBCO(3ML) in the $z(xy)\bar{z}$ geometry at 60 and 10 K.

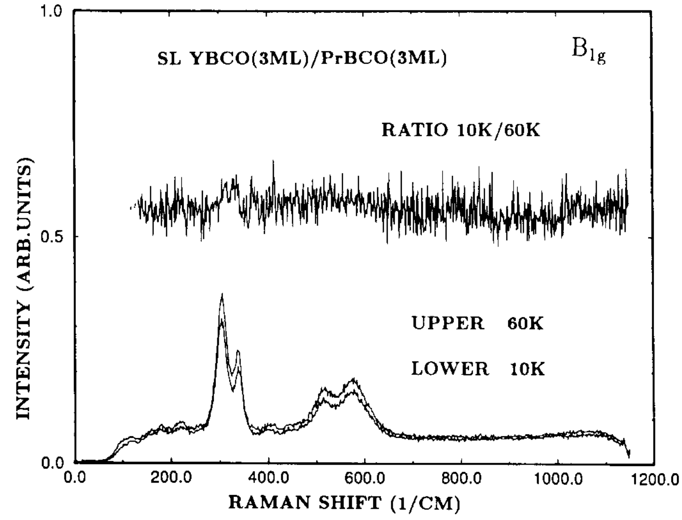

Figure 5: Raman spectra of the SL YBCO(3ML)/ PrBCO(3ML) in the $z(xx)\bar{z}$ geometry at 60 and 10 K.

4 Far-infrared reflectivity

Infrared studies of HTSC SL have been motivated by a desire to determine the energy gap and also the carrier concentration in the YBCO layers. Figure 6 shows reflectivity spectra measured at room temperature for five different samples. While the 4000Åthick YBCO film has a featureless spectrum with reflectance above 90% [10], the PrBCO spectrum exhibit a rich structure due to phonons which is typical for dielectrics. The spectrum of the SL

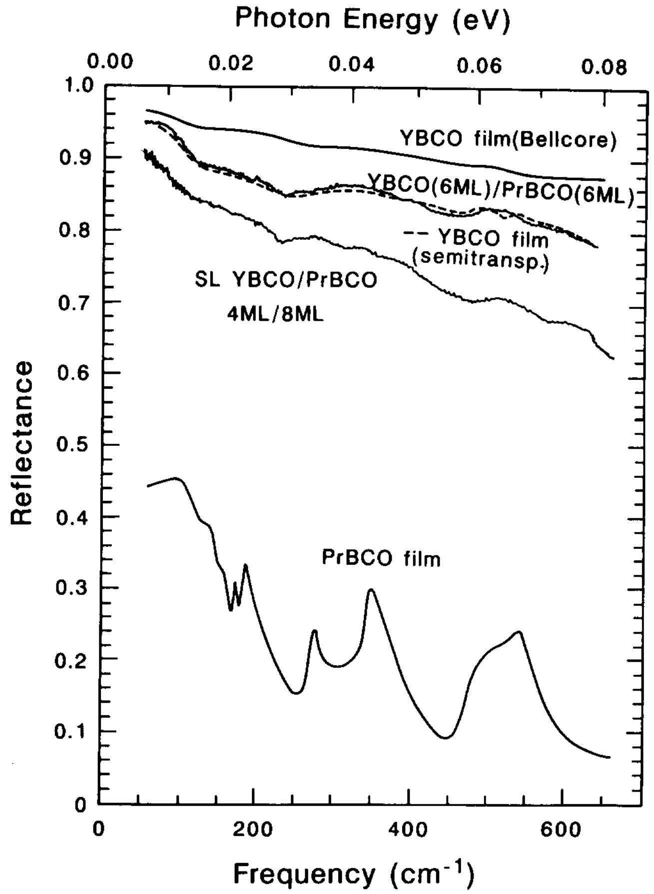

Figure 6: FIR reflectance of thick and semitransparant YBCO films, PrBCO film, YBCO(6ML)/PrBCO(6ML) and YBCO(4ML)/PrBCO(8ML) superlattices.

YBCO(6ML)/PrBCO(6ML)×40 periods (5760Å) with T_c=83K resembles very much that of the pure YBCO. Some decrease of the reflectance is reminiscent of the substrate (compare to the 3000Å/thick YBCO film with T_c=91K). It is important to note that PrBCO phonons are nearly completely screened in the SL, which may be evidence of a strong interlayer interaction. It is clearly seen that the reflectivity spectrum of the SL YBCO(4ML)/PrBCO(8ML) (T_c=52K) decreases more rapidly as a function of frequency. Such a decrease is similar to that observed in YBCO crystals with T_c=50K [11]. The changes observed in the spectra may be interpreted in terms of carrier depletion in YBCO layers.

5 Conclusion

The experimental results described in this paper demonstrate that an YBCO-based SL provide us with a powerful tool for investigating the fundamentals o the high-temterature superconductivity phenomenon. The systematic variation of T_c in SL, although not completly understood, allows the possibility to find correlations between Raman and infrared spectra and transport properties of these materials.

6 References

1. U. Poppe, P. Prieto, F. Schubert, H. Saltuer, K. Urban, Solid State Commun. **71**, 569 (1989).
2. J.-M.Triscone, M.G.Karkut, L.Antognazza, O.Brunner, and Ø.Fischer, Phys.Rev.Lett. **63**, 1016 (1989).
3. Q.Li, X.X.Xi, X.D.Wu, A.Inam, S.Vadlamannti, W.L.McLean, T.Venkatesan, R.Ramesh, D.M.Hwang, J.A.Martinez, and L.Nazar, Phys.Rev.Lett. **64**, 3086 (1990).
4. R.F.Wood, Phys.Rev.Lett.**66**, 829,(1991)
5. C.Timusk and D.B.Tanner, in *Physical Properties of High-Temperature Superconductors III*, ed. by D.M. Ginsberg (World Scientific, Singapore, 1992), p.363.
6. C.Thomsen and M.Cardona, in *Physical Properties of High-Temperature Superconductors I*, ed. by D.M. Ginsberg (World Scientific, Singapore, 1989), p.407.
7. A.P.Litvinchuk, C.Thomsen, I.E.Trofimov, H.-U.Habermeier, and M.Cardona, Phys. Rev. **B46** (1992), in press.
8. C.Thomsen, R.Wegerer, H.-U.Habermeier, and M.Cardona, Solid State Comm. **83**, 199 (1992).
9. S.L.Cooper,M.V.Klein, B.G.Pazol, J.P.Rice, and D.M.Ginsberg, Phys. Rev. **B37**, 5920 (1988).
10.K.Kamarás, S.L.Herr, C.D.Porter, N.Tache, D.B.Tanner, S.Etemad, T.Venkatesan, E.Chace, A.Inam,X.D.Wu, M.S.Hegde, and B.Dutta, Phys.Rev. Lett. **64**, 84 (1990)
11. J.Orenstein, G.A.Thomas, A.J.Millis, S.L.Cooper. D.H.Rapkine, T.Timusk, L.F.Shneemeyer, and J.V.Waszczak, Phys. Rev. **B42**, 6342 (1990)