Elastic constants of metal-insulator superlattices

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A Brillouin scattering study of elastic properties of NbN/AlN superlattices is presented. Because the coupling of light to surface waves in this system is very weak, we describe a modification of the technique which enhances the scattering efficiency, thereby allowing measurements of a system which is otherwise inaccessible. This particular multilayer superlattice is one of the very few that does not exhibit any elastic anomalies as a function of layer thickness in accordance with the idea that electron transfer may be the mechanism responsible for elastic anomalies in superlattices.

The measurement of elastic constants of thin films and multilayers is of current interest due to expectations that novel materials can be prepared by artificially growing them in thin-film geometry. The elastic properties of metallic superlattices have received considerable attention due to the discovery of anomalous behavior in a large number of systems.[1] Many electronic devices are built in thin-film form; therefore, their behavior and performance are strongly affected by modifications that occur in thin films. In particular, the mechanical properties of films and substrates are of practical importance because most applications require rugged, strongly bonded devices.

The study of the mechanical properties of thin films can be performed using a variety of techniques which require removing the films from the substrates.[1] This is quite cumbersome and undesirable particularly because the possibility exists that the elastic properties of the films may change when removed from the substrate. Brillouin scattering has been shown to be a useful technique for the study of many thin films on substrates.[2,3] Even in the case of metallic films, where the small penetration depth of light precludes coupling through the conventional elasto-optic mechanism, Brillouin scattering from surface waves has yielded information on elastic properties. In this latter case, the coupling takes place by means of the ripple mechanism[2] which depends on the corrugation of the surface produced by the phonon and on the value of the dielectric constant of the material. Cases exist, however, in which the dielectric constant of a film is such that the coupling to the surface modes is very weak, thereby making the use of Brillouin scattering impossible. Here we describe a way to overcome this difficulty so that Brillouin scattering can be used to study materials that otherwise show no Brillouin signal. We have applied this technique to the study of elastic constants of the metal/insulator (NbN/AlN) superlattice. Many superlattices (e.g., Cu/Ni, Nb/Cu, Mo/Ta, V/Ni, Au/Cr, etc.) have been shown to exhibit a marked dependence of their elastic properties on the modulation wavelength (Λ);[1] this behavior is contrary to that predicted by conventional elasticity theory which predicts that elastic properties should be independent of Λ. We show that the elastic anomalies generally observed in metal/metal superlattices are not present in NbN/AlN in accordance with the idea that electron transfer[4] may be responsible for anomalies in the structure and elastic constants.

High quality NbN/AlN multilayers were grown on sapphire substrates held at 200 °C using a sputtering technique described earlier.[5] Briefly, the samples are deposited on substrates which are alternately moved using a computer-controlled stepping motor between two rate-stabilized sputtering guns. Because the concentration of nitrogen in NbN and AlN is known to strongly affect their properties, the reactive sputtering was accomplished while controlling the N pressure in the sputtering gas using a mass spectrometer in a feedback mode.[5] The samples were characterized using high- and low-angle x-ray diffraction[6] together with microcleavage transmission electron microscopy.[7] Brillouin scattering measurements were performed on a tandem ($5+2$ pass) Fabry–Perot interferometer.[2]

As-grown NbN, AlN, or NbN/AlN multilayers show no noticeable Brillouin scattering. This result is not surprising for AlN which, because it is an insulator, must have a relatively small dielectric constant and consequently a low reflectivity. For NbN, on the other hand, the general rule that highly reflecting metallic surfaces produce strong signals does not hold; we assume this to be due to some particular combination of real and imaginary components of the dielectric tensor. In order to enhance the light scattering cross section we evaporated a thin (~ 150 Å) layer of Al (which is known to produce a strong signal[2]) on each sample; in this fashion Brillouin spectra were obtained from all samples. Two spectra taken with and without the Al film are shown in Fig. 1. The lower spectrum is from a sample *without* an Al film which was accumulated for 53 min; the upper spectrum (offset vertically by 5 counts/channel) was from another sample *with* an Al film which was accumulated only for 18 min. The peaks labeled B are the Brillouin peaks and those labeled with an asterisk are of instrumental origin. Since the phonon wavelengths probed in these types of measurements are approximately 3000 Å, the effect of 150 Å of Al at the surface is not negligible. Because the velocity in pure Al is lower than that of NbN/AlN multilayers, all measured values will be a few percent lower than for an uncoated sample. For a meaningful comparison between samples this

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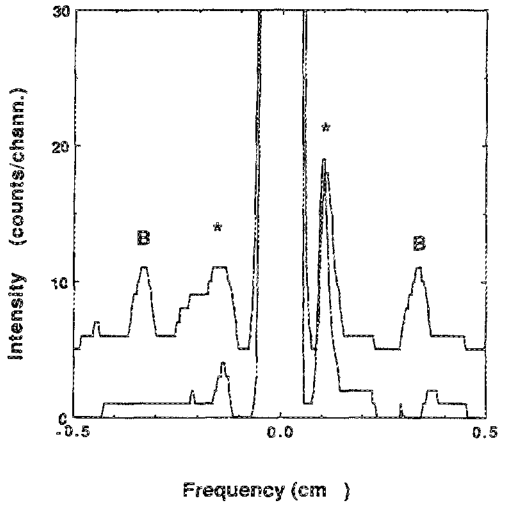

FIG. 1. Spectra from NbN/AlN multilayers. The lower spectrum was from a sample with no Al layer at the surface and ran for 53 min. The upper spectrum (offset by 5 counts/channel) was from a sample with a 150 Å Al coating and ran for 18 min. The symbols *B* and * indicate Brillouin lines and peaks of instrumental origin, respectively.

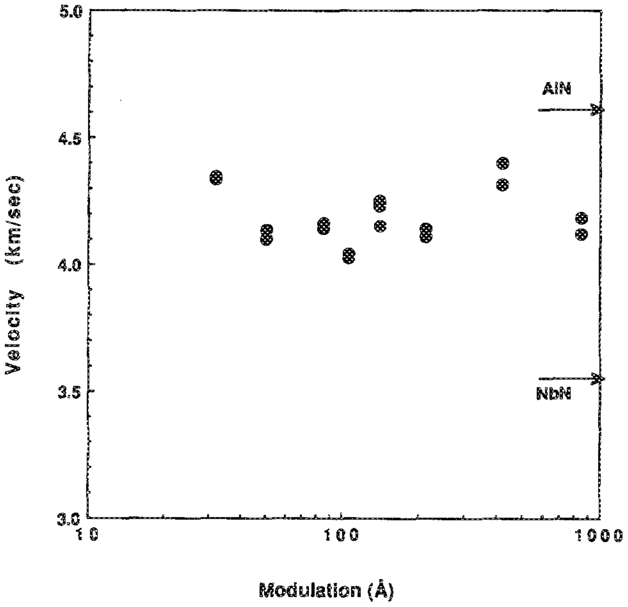

FIG. 2. Modulation wavelength dependence of surface wave velocity in AlN/NbN superlattices with equal thicknesses of AlN and NbN layers. The arrows indicate the surface wave velocity in pure AlN and NbN films. (All films were capped by a 150 Å Al layer to enhance the coupling to the radiation.) Each dot represents a different run; for a given sample the spread in data points is representative of the experimental accuracy.

change must be the same for all samples. To avoid uncertainties arising from changes in the thickness of the Al films on each sample, we deposited the Al film on all samples simultaneously. These simple modifications allow Brillouin scattering to be used to study many thin-film systems that were previously inaccessible. The only conditions necessary for this technique to be applicable is that the overlayer should couple to the light and adhere well to the underlying material whose elastic properties are being investigated.

Now we turn our attention to the specific system under study, NbN/AlN superlattices. It was shown earlier[1] that most metal-metal multilayers exhibit some type of elastic anomaly for layer thicknesses around 20 Å. For cases in which structural data are also available, this anomaly was shown to be correlated with a lattice expansion or contraction.[8,9] Numerical simulation studies[8] showed that the lattice expansion is able to explain the elastic anomalies observed in Mo/Ni; a phenomenological model based on Murnaghan's equation state[9] was also able to explain the elastic anomalies in a number of systems as due to the observed lattice changes. The origin of the expansion, however, was not identified in these investigations.

A number of possible explanations to account for the elastic behavior of superlattices have been proposed and can be broadly classified into two categories: structural and electronic. In the former category recent calculations[10] have shown that the presence of grain boundaries is capable of explaining some of the observed anomalies. Questions as to whether this model can explain not only softenings but also the experimentally observed hardenings are sill awaiting investigation. Early explanations based on electronic effects[11-13] are similar to the Hume–Rothery effect[14]; i.e., when the new Brillouin zone created by the superlattice touches the Fermi surface, an anomaly develops in the dielectric function and consequently also in the elastic constants. However, all superlattices studied to date exhibit large interfacial electronic scattering (as observed in transport measurements[15,16]) which excludes the existence of extended electronic states. This type of effect is therefore unlikely to be the driving mechanism in most cases.

It has been conjectured[8] that the observed lattice expansion may be due to electron transfer and one of us recently correlated the size of the expansion with differences in work functions.[4] In order to ascertain if this approach is indeed a valid one it is important to study a wide range of materials with different amounts of electron transfer. NbN/AlN is a system formed from a metal (NbN) with a wide gap semiconductor (AlN) which is therefore expected to exhibit very little electron transfer, and consequently the elastic constants are not expected to change significantly as a function of modulation wavelength.

Figure 2 shows the measured phonon velocity (v) as a function of modulation wavelength for a series of NbN/AlN equal thickness multilayers. Within the experimental error, the phonon velocity is constant as expected[4] from a sample which exhibits no electron transfer. We conclude from our measurements that in NbN/AlN any elastic anomaly is less than (8%) which is to be compared with changes of up to 50% in metal-metal superlattices.

In summary, we have demonstrated a technique which allows the application of Brillouin scattering to systems which do not otherwise couple to light. This technique was applied to the measurement of the elastic constants of NbN/AlN. In accordance with expectations based on electron transfer arguments, NbN/AlN shows no changes in the elastic constants. Further experimental work as well as comparisons with detailed quantitative theoretical calculations is necessary before the origin of elastic anomalies in superlattices can be uniquely ascertained.

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