Optical Investigation of ultra-thin NbN films for phonon-cooled Hot-Electron-Bolometers (HEB)

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Abstract— **We investigate ultra-thin NbN films by means of spectroscopic ellipsometry and Raman spectroscopy. These measurements give important information on thickness dependent structural properties and phonon density of states. Thus first steps towards a microscopic understanding of the physics of phonons in these films and their coupling to the substrate can be made. As an ultimate goal we search to relate optical measurements with the specific performance of HEBs and in particular the IF bandwidth of HEB mixers. This work is part of a larger project to optimize NbN films for HEB applications including a relatively large number of deposition parameters. Within this parameter study a first surprising result is that a native Si <100> surface is non-optimal for good superconductive properties of NbN films.**

***Index Terms*—Hot-Electron-Bolometer (HEB), NbN, Raman-spectroscopy, superconducting properties**

I. INTRODUCTION

HEB mixer devices based on superconducting NbN films have been manufactured by various groups [1,3,4,5,6,7, 15,16,17,18,19,20]. One of the main limitations of HEB mixers for THz spectroscopy remains the relatively small IF bandwidth (~3-4 GHz) resulting in an exceedingly small Doppler-velocity range for astronomical applications. According to current understanding, the IF bandwidth of phonon-cooled HEBs is largely determined by the time scales on which phonons escape from the thin film into the substrate. Apart from film thickness, sound speed and acoustic matching between film and substrate play therefore a major role. Unfortunately, so far only very little is known about these parameters and most often these parameters have been freely adjusted to more complex measurements or have been derived from bulk values. To optimize films and devices it is however necessary to develop a detailed microscopic picture of the phonon escape process. On one side this requires modeling which takes into account the particular situation of phonons in very thin films. On the other side it is mandatory to search for measurements which allow direct access to the structure and dynamics of films with thickness below 5 nm. Due to the relative strong absorption of light in NbN films, optical investigations are surface sensitive. In this paper we present first results of spectral ellipsometry and Raman scattering and discuss preliminary interpretations of these measurements.

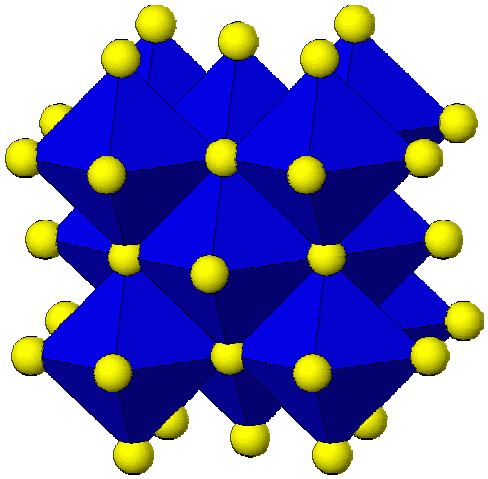


Fig.1. Lattice structure of an ideal fcc NbN single crystalline film. The spheres correspond to the nitrogen atoms. The niobium atoms are located in the center of the bi-pyramids. We found that the Raman-scattering cross-section of this material is sufficient for investigations of ultra-thin films.

II. FILM PREPARATION AND PARAMETER SPACE

In the standard process of IRAM, thin NbN films are deposited from a 4 inch niobium target by 13.56 MHz RF magnetron sputtering technique on a 2 inch substrate in a nitrogen / argon / methane atmosphere (pressure: 0.852 Pa, flow Ar: 46 sccm, flow N_2: 2.7 sccm, flow CH_4: 0.6 sccm, power 240 W). The deposition is made at room temperature. The small amount of methane improves the superconducting properties of the NbN film [6,7]. However, carbon build into the films by this process might influence the device performance.
As a first approach before a more general parameter study we try to understand the thickness dependence of optical and electrical properties of the films.

The dependence of the optical constants n and k at room temperature of NbN and the superconducting transition temperature on thickness and on partial methane pressure during the deposition will be discussed in the following chapters. A first attempt to use silicon as appropriate substrate material will be discussed.

III. Spectral Ellipsometry and Raman Spectroscopy

The evaluation of the phonon cooling mechanism by an acoustic matching approach using the long wavelength properties as derived from bulk materials might not be optimal for ultra-thin NbN films. These films have usually polycrystalline or amorphous lattice structure [6,9]. Film stress and lattice disturbances can lead to important variations in the acoustic properties and the very small NbN film thickness will lead to changes in the phonon density of states.

Fig. 2 shows wavelength-dependent ellipsometric measurements of the optical constants n and k of ultra-thin NbN films on fused quartz with 15 nm MgO seed layer [13]. An indication that important, thickness-dependent structural and electronic evolution is taking place is given by the thickness-dependent complex refractive index. For a higher film thickness a shift of the minimum in the refractive index n down to lower wavelengths λ can be observed.

NbN is Raman-active. For NbN films the wavelength dependent Raman scattering depth δ_R is entirely dominated by the strong absorption :

$$\delta_R(\lambda) = \frac{1}{2} \cdot \frac{\lambda}{4\pi \cdot k(\lambda) \cdot n(\lambda)} \qquad (1)$$

Raman scattering measurements of NbN are therefore surface sensitive. For the wavelength of He-Ne laser (λ = 632.817 nm) the corresponding scattering depth is about $\delta_R \approx 8$ nm.
We investigated NbN films by confocal Raman scattering setup with a spot diameter of 1 μm. To avoid heat-related oxidation effects of NbN in air we used a low laser power of P = 6.5 mW.

First Raman measurements have been reported in [1,11]. Fig. 3 shows the Raman-spectra of ultra-thin NbN films of different thickness as measured at room temperature. The right panel illustrates the corresponding bulk NbN phonon dispersion relation between Γ and X as measured by neutron diffraction methods [12]. A first information about the film structure is given by the very strong signal from acoustic phonons. This indicates a violation of the momentum conservation due to grain sizes in the nm range. Former X-ray diffraction measurements show similar grain sizes [7,9]. For such polycrystalline films the Raman signal form of acoustic phonons is a good indicator of the phonon density of state (DOS).

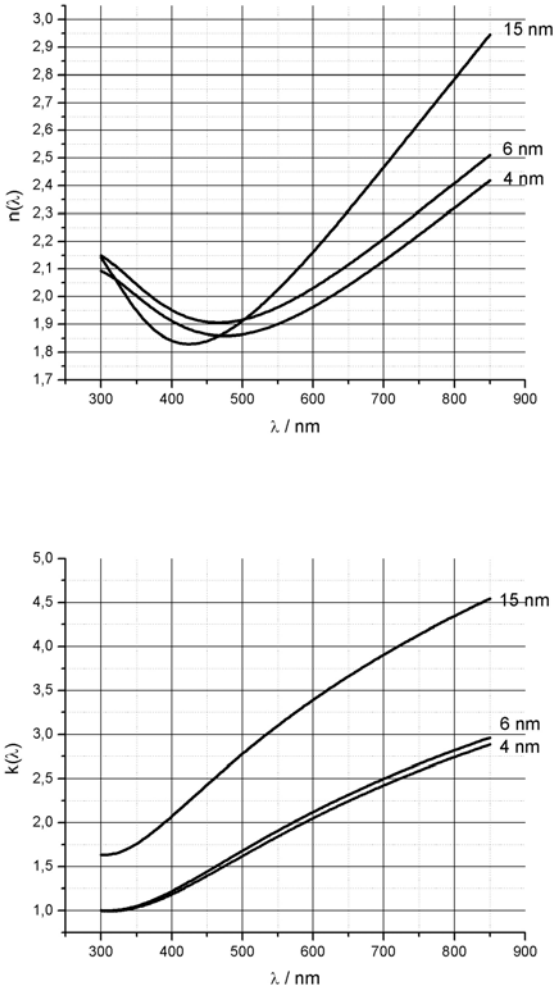


Fig. 2. Wavelength dependent ellipsometric measurements of the optical constants n and k of ultra-thin NbN films on fused quartz with a 15 nm MgO seed layer.

For films of a thickness less than 3 nm a shift to higher frequencies and simultaneously a considerable broadening of the acoustic DOS peak can be observed. This effect can be either due to stress or phonon confinement.

IV. Superconducting Properties

In a second step we have studied the superconducting properties of reactive sputtered ultra-thin NbN films. An important parameter is the critical temperature T_c of the NbN film obtained by DC transport measurements. We investigated the dependence of the resistivity R(T) as function of the temperature T for various film thickness and for different lattice-stabilizing methane contents in the sputter gas atmosphere during the deposition process. The dependencies are shown in Fig. 4.

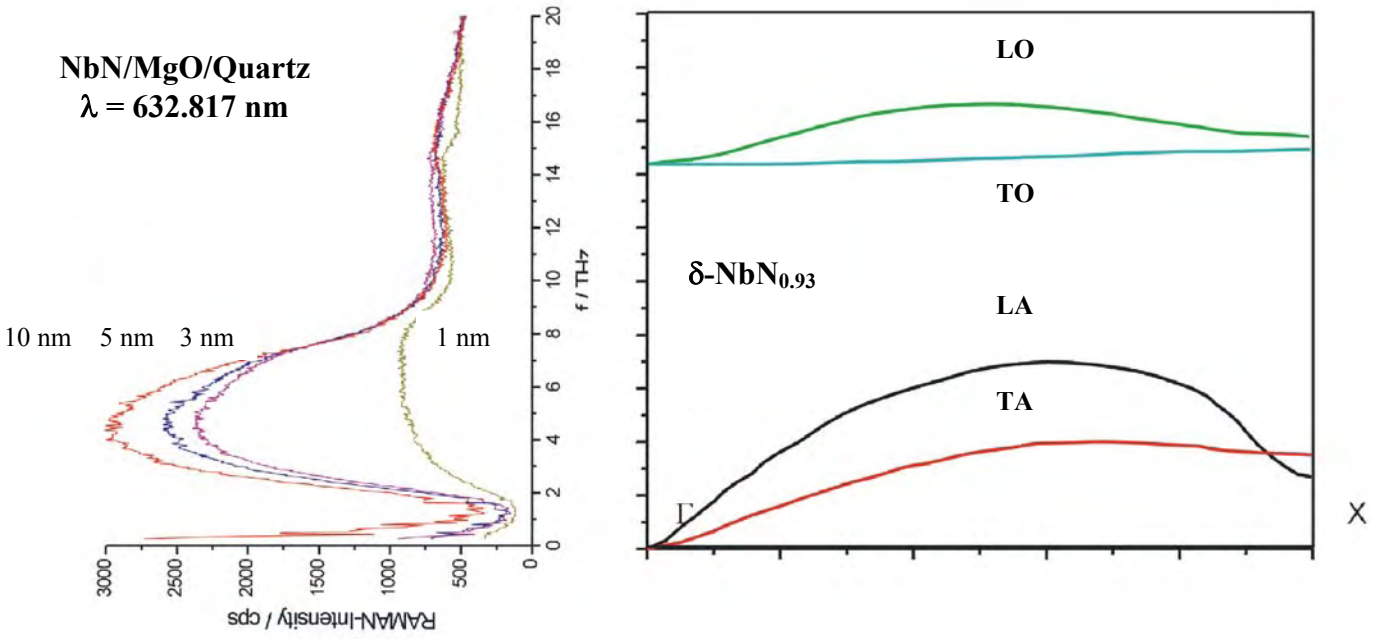
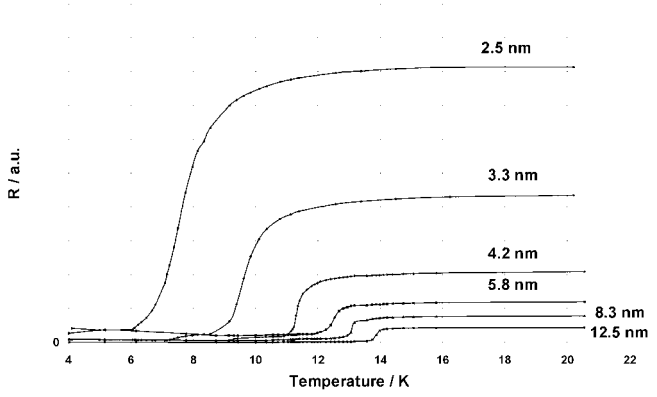


Fig. 3. Raman-spectra of ultra-thin NbN films and the corresponding NbN phonon-dispersion relation.

Real HEB devices use a NbN film thickness between 3.5 nm and 5 nm. The critical temperature in this range is about Tc ≈ 9 – 11 K. The standard methane flow in the IRAM deposition process is 0.6 sccm CH_4 for a gas mixture of 46 sccm argon and 2.7 sccm nitrogen. The total deposition pressure is p = 0.85 Pa. For a higher methane flow the critical superconducting temperatur is increased.

To improve the thermal properties of HEB devices we take into account different substrate materials. A first attempt is the use of (100)-orientated silicon substrates. The surface of pure silicon is covered by a native SiO_x layer [14]. The quality of reactive sputtered NbN on silicon substrates shows a dependence of the resistivity R(T) from the cleaning process of an argon plasma, illustrated in Fig. 5.



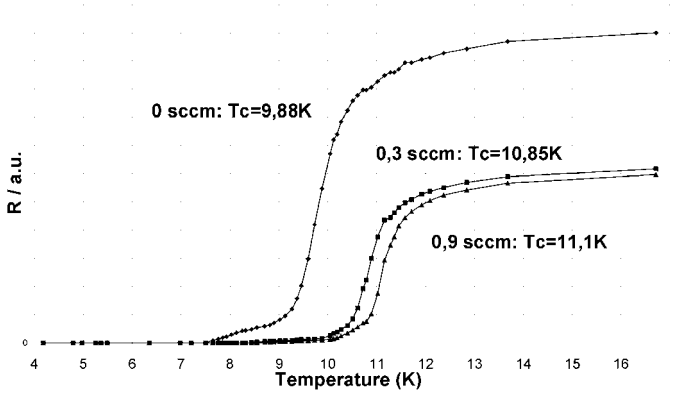


Fig. 4. Dependence of the resistivity R(T) of ultra-thin NbN films on fused quartz with 15 nm MgO seed layer for different thickness and lattice carbon content. The thickness of the NbN film is about 4 nm.

It is remarkable that without cleaning the resistivity is lower than with cleaning procedure.

V. Conclusion

Optical investigations by means of ellipsometry and Raman spectroscopy of ultra-thin NbN films can reveal information on structural and electronic properties. Our measurements indicate important interface stress for NbN films as grown on MgO buffer layers.

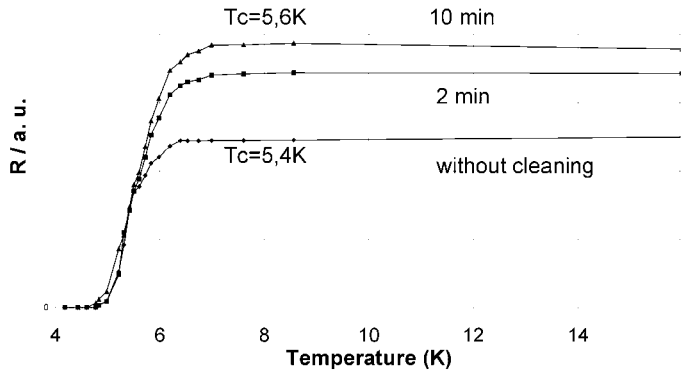


Fig. 5. Temperature dependence R(T) of 4 nm thick NbN films on (100)-oriented silicon substrate without MgO seed layer with different argon plasma cleaning time. The plasma power was 50 W (13.56 MHz RF) at 2 Pa (50 sccm) Ar pressure. The power density for the cleaning process is about 2.5 W/cm^2.

Ellipsometry indicates a thickness dependence of refractive index which is likely due to structural inhomogeneity. Films on clean Si <100> show reduced superconducting properties as compared to Si substrates with native oxide, an effect which is likely due to the large lattice mismatch between NbN and Si.

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