Optical investigations of the collective transport in CDW films

Artem V. Pronin, Martin Dressel, Alois Loidl, H. S. J. van der Zant, O. C. Mantel, C. Dekker

Angaben zur Veröffentlichung / Publication details:

Pronin, Artem V., Martin Dressel, Alois Loidl, H. S. J. van der Zant, O. C. Mantel, and C. Dekker. 1998. "Optical investigations of the collective transport in CDW films." *Physica B* 244 (1): 103–6. https://doi.org/10.1016/S0921-4526(97)00470-5.

Optical investigations of the collective transport in CDW-films

A.V. Pronin[a,1], M. Dressel[a,*], A. Loidl[a], H.S.J. van der Zant[b], O.C. Mantel[b], C. Dekker[b]

[a] *Experimentalphysik V, Universität Augsburg, Universitätsstr. 1, D-86150 Augsburg, Germany*
[b] *Department of Applied Physics and DIMES, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands*

During the last 25 years the electrodynamic properties of many materials which exhibit a charge-density-wave (CDW) ground state were intensively studied by different techniques in order to obtain information on the frequency dependence of the complex conductivity [1, 2]. The group of Wachter [3, 4] first found the opening of a single-particle gap in the excitation spectrum of $K_{0.3}MoO_3$ (blue bronze) by means of infrared spectroscopy. The second important feature of such materials is the appearance of a collective mode at finite frequencies which is associated to the CDW pinned by impurities. For most of the CDW-compounds like $NbSe_3$, TaS_3, or $(TaSe)_4I_2$ this resonance is located in the microwave or radio frequency range [1, 2] and can be studied by standard high-frequency techniques. In other CDW-materials such as molebdenium bronzes ($K_{0.3}MoO_3$, $Rb_{0.3}MoO_3$, etc.) this resonance appears in the millimeter and submillimeter frequency range (1–10 cm^{-1}) and only few experimental data are available [5–7]. Since $R \approx 1$, the accuracy of reflectivity measurements at this regime is not sufficient for reliable conclusions concerning quantitative estimations of the conductivity behavior. The availability of thin blue-bronze films [8] allowed us to perform the transmission measurements and to continuously measure the real and imaginary parts of the conductivity in the frequency range of interest. This kind of transmission measurement on thin films has previously proven to be a very powerful method for studying the electrodynamical properties of highly conducting systems [9–11].

* Corresponding author.

[1] On leave from General Physics Institute, Russian Academy of Sciences, Moscow, Russia.

In our experiments, we used two thin films of the charge-density-wave compound $Rb_{0.3}MoO_3$ with thicknesses of 250 and 750 nm, respectively. The films have been grown by pulsed-laser deposition on a plane-parallel 5×5 mm sapphire substrate with orientation (0 1 2) and thickness of about 0.45 mm. The single-phase films consisted of grains with typical sizes of the order of micrometers. X-ray analysis indicates that the *b*-axis along which the CDW develops is oriented in the plane and most of the grains are single crystalline. Although the grains can be well aligned along one direction, the films used in this study were oriented in the plane to a small degree only. Details of the film preparations and characterization are given in Ref. [8].

The measurements in the millimeter frequency range (3–9 cm^{-1}) were made with a coherent-source spectrometer in quasi-optical configuration [12]. Three different backward-wave oscillators were used as continuously tunable sources of monochromatic and linearly polarized radiation. A Mach–Zehnder interferometer arrangement allows us to measure both the transmission coefficient and phase shift. The samples were cooled down from room temperature to 3 K. The measurement of a bare Al_2O_3 substrate of the same (0 1 2) orientation were performed to obtain the optical parameters of the substrate. The conductivity and the dielectric constant of the films were directly calculated for each frequency point from the transmission and phase-shift measurements by using the Fresnel's formulae for a two-layer system [13]. For further discussion, we concentrate on the thicker film, since the main experimental features and results obtained are the same for the film with 250 nm thickness.

In Fig. 1, the original data of a transmission and phase-shift spectra obtained at $T = 100$ K are displayed for the 750 nm film on the Al_2O_3 substrate. Two large oscillations in the transmission spectra are due to interference inside the substrate which acts as a Fabry-Perot resonator. The period of the oscillations is mainly controlled by the refraction index n_s and the thickness d_s of the substrate. The level of the transmission is defined by the transparency of the $Rb_{0.3}MoO_3$ film, which is governed by its conductivity. The frequency slope of the phase oscillations is determined by the film properties.

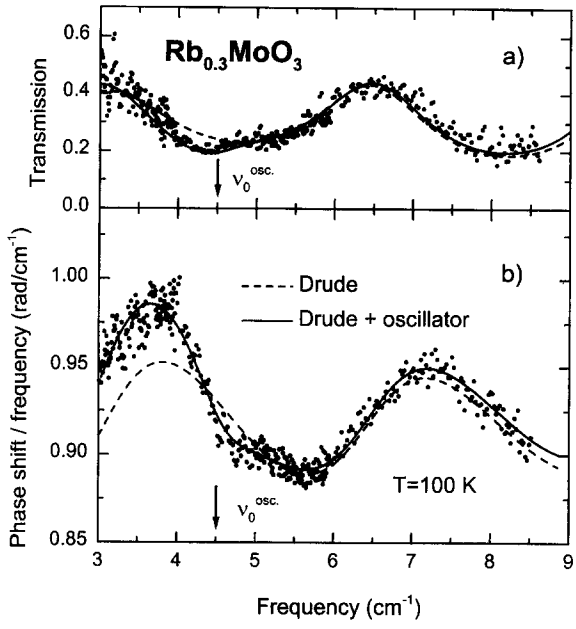

Fig. 1. Experimental data of the transmission (a) and the phase shift (b) spectra of a 750 nm $Rb_{0.3}MoO_3$ film on a 0.45 mm Al_2O_3 substrate obtained at 100 K. The dashed lines represent fits by the Drude model; the solid lines correspond to fits by a Drude model and an oscillator. (The phase shift is divided by frequency for clarity.)

The points in Figs. 2 and 3 present direct calculations of the frequency dependence of conductivity and dielectric constant of the film for several temperatures above and below the Peierls transition temperature ($T_{CDW} = 180$ K). For the temperatures below T_{CDW} in the spectra of σ and ε' we find clear evidence for a weak but a vivid mode centered at 4.5 cm^{-1} and with a line width of about 1 cm^{-1}.

The evaluation of the mode is done by fitting the original transmission and phase-shift spectra using the simplest possible model consisting of a Drude term and a single Lorenz oscillator. In the example of such fits, shown in Fig. 1, it is clearly seen that the oscillator term is crucial to model the experimental results. The frequency-dependent conductivity and dielectric constant of $Rb_{0.3}MoO_3$ obtained by these fits are represented in Figs. 2 and 3 by solid lines. We found, that the Drude conductivity drops from about 90 $\Omega\ cm^{-1}$ at 150 K down to almost zero at 6 K (Fig. 4), in agreement with the DC conductivity. The mode described by a Lorentz

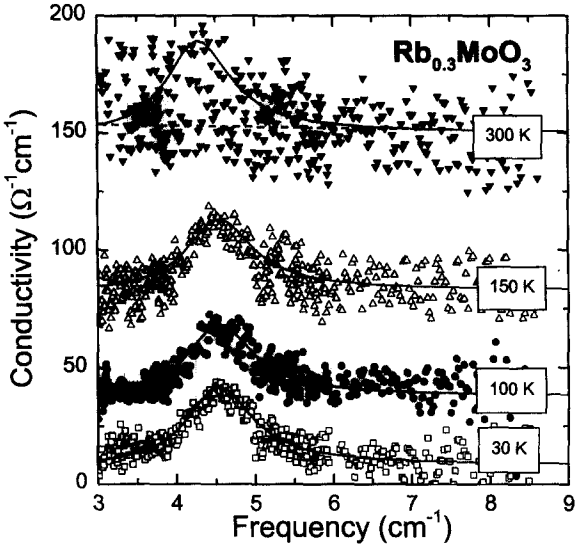

Fig. 2. Frequency-dependent conductivity of the $Rb_{0.3}MoO_3$ film for different temperatures above and below $T_{CDW} = 180$ K as indicated; the curves are *not* shifted. The points represent results directly calculated from the transmission and phase-shift spectra. The solid lines correspond to the fits of the transmission and phase-shift spectra. The dashed line shows a fit of the 300 K conductivity data by a single Drude term.

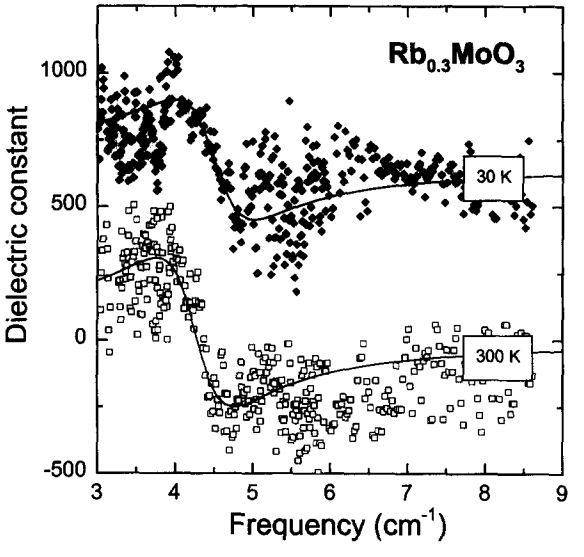

Fig. 3. Frequency dependence of the dielectric constant of the $Rb_{0.3}MoO_3$ film at $T = 30$ and 300 K. All spectra at temperatures below T_{CDW} practically coincide with the 30 K data. The points represent directly calculated results. The solid line is obtained by fits of the transmission and phase-shift spectra.

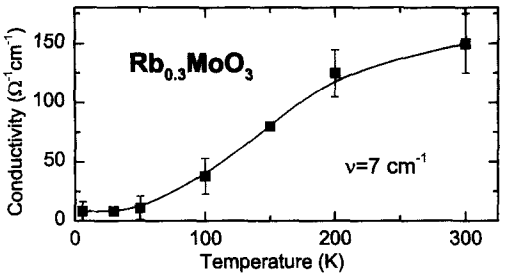

Fig. 4. Temperature dependence of the Drude conductivity of the $Rb_{0.3}MoO_3$ film; the solid line is a guide to the eye.

oscillator (center frequency $\nu_0 = 4.5\ cm^{-1}$, scattering rate $\gamma = 1\ cm^{-1}$, oscillator strength $\nu_p = 45\ cm^{-1}$, leading to $\Delta\varepsilon = \nu_p^2/\nu_0^2 = 100$) remains practically unchanged (within our experimental accuracy) for temperatures below T_{CDW}. Due to the decreasing conductivity of the broad background, this feature becomes more pronounced as the temperature decreases. We attribute this mode to the collective excitations of the charge-density-wave condensate pinned to impurities. Microwave cavity measurements on bulk samples of $K_{0.3}MoO_3$ [6] and estimations from the threshold field of the non-linear conductivity [1, 2] infer that the pinned-mode resonance is located around $3.3\ cm^{-1}$. We explain the slightly higher pinning frequency observed in our experiments on thin films with the higher defect concentration compared to single crystals and the finite grain size of typically 1 μm. The thickness of the film has only little influence since we observe the same resonance frequency in both samples (750 and 250 nm). We feel that the width of the mode is due to a distribution of resonance frequencies rather than homogeneous broadening.

Our measurements of $Rb_{0.3}MoO_3$ films allowed us to estimate the spectral weight of the collective mode and, consequently, the effective mass m^* of the condensate. Taking the single-particle plasma frequency from infrared measurements [6], we obtain $m^*/m_b \approx 10^4$, which is somewhat higher than estimated from previous measurements of $K_{0.3}MoO_3$ [5–7]. Since the films were not single crystalline but a mixture of randomly oriented grains in the plane, the polarized radiation effectively interacts with half of the CDWs developing

in the grains along the *b*-direction. We can not also exclude that some part of the film is non-stochiometric or amorphous.

We can compare our experimental results with data for the gap energy $\hbar\omega_{gap}$ and the plasma frequency ω_p of blue bronze, using the standard formula [14] for the increase of the dielectric constant due to the opening of the single-particle gap: $\Delta\varepsilon = 2\omega_p^2/3\omega_{gap}^2$. The estimations found in literature [6] give values for $\Delta\varepsilon$ of about 10^2, which is in agreement with the rise in the dielectric constant we observe (Fig. 3) as the temperature is lowered; this behavior therefore indicates the opening of the single-particle gap at approximately 10^3 cm^{-1}.

Now, let us consider the dynamical conductivity and dielectric constant of $Rb_{0.3}MoO_3$ in the normal state, above T_{CDW}. Spectra of the conductivity in this case are noisy due to the low transmission of the sample. From the conductivity data obtained at room temperature the existence of mode around 4.5 cm^{-1} cannot be definitely implied, as shown by the dashed line (only Drude model) and the solid line (Drude-plus-Lorentz model) in Fig. 2. The dispersion of the dielectric constant around 4 cm^{-1} (Fig. 3), however, gives clear evidence of the existence of a collective excitation even at temperatures above T_{CDW}. This feature is related to fluctuations of the CDW interacting with impurities; indications of this behavior have been observed previously [15, 16].

Summarizing, the complex dynamic response of thin $Rb_{0.3}MoO_3$ films was studied by means of millimeter–submillimeter spectroscopy technique. In the spectra of conductivity and dielectric function at temperatures below T_{CDW} we have observed the characteristic features of the CDW-pinned mode centered at $\nu_0 = 4.5$ cm^{-1}. The parameters of this mode are practically temperature independent. Above T_{CDW} we have found indications of CDW fluctuations even at room temperature.

We would like to thank G. Grüner for helpful discussions. Part of the work was supported by the Russian Foundation for Basic Research and by the Deutsche Forschungsgemeinschaft through the German-Russian scientific cooperation program.

References

[1] G. Grüner, Rev. Mod. Phys. 60 (1988) 1129.
[2] G. Grüner, Density Waves in Solids, Addison-Wesley, Reading, MA, 1994.
[3] G. Travaglini et al., J. Phys. Soc. Japan 49 (Suppl. A) (1980) 869.
[4] G. Travaglini et al., Solid State Commun. 37 (1981) 599.
[5] H.K. Ng et al., Phys. Rev. B 33 (1986) 8755.
[6] L. Degiorgi et al., Phys. Rev. B 44 (1991) 7808.
[7] L. Degiorgi, G. Grüner, Phys. Rev. B 44 (1991) 7820.
[8] O.C. Mantel et al., Phys. Rev. B 55 (1997) 4817.
[9] B.P. Gorshunov et al., Solid State Commun. 87 (1993) 17.
[10] A.V. Pronin et al., Sov. Phys. JETP 82 (1996) 790.
[11] M. Dressel et al., These Proceedings.
[12] A.A. Volkov et al., Infrared Phys. 25 (1985) 369.
[13] M. Born, E. Wolf, Principles of Optics, Pergamon Press, Oxford, 1980.
[14] P.A. Lee et al., Solid State Commun. 14 (1974) 703.
[15] B.P. Gorshunov et al., Phys. Rev. Lett. 73 (1994) 308.
[16] A. Schwartz et al., Phys. Rev. B 52 (1995) 5643.