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RAYLEIGH SCATTERING OF MOSSBAUER RADIATION ON VAPOUR-QUENCHED Te*

F.M. Wagner, F.J. Litterst and G.M. Kalvius

Physik Department der Technischen Universität München, D-8046 Garching, Germany.

Résumé. - Du tellure non cristallin a été préparé par déposition d’un faisceau moléculaire sur un substrat de mylar maintenu à 80K. La structure amorphe est fortement influencée par l’épaisseur de l’échantillon. Des échantillons minces (<10nm) ont été chauffés à partir de 80K à une vitesse de ~15K/jour. A 245K on observe une diminution brusque de l’intensité de diffusion Rayleigh élastique. Ceci correspond à une augmentation de l’amplitude moyenne de vibration atomique \( \sqrt{x^2} \) qui passe de ~0.15A à ~0.32A. Après avoir maintenu l’échantillon pendant 10 heures aux alentours de 245K, on retrouve la valeur initiale de \( \sqrt{x^2} \). Les spectres de RX montrent que cette variation de \( \sqrt{x^2} \) est liée à un réarrangement structural. Dans le même intervalle de température, des échantillons épais cristallisent et ne présentent que de faibles anomalies de \( \sqrt{x^2} \).

Abstract. - Non-crystalline Te was prepared by deposition from a molecular beam onto a mylar substrate kept at 80K. The amorphous structure is strongly influenced by sample thickness. Thin samples (<10nm) were heated from 80K at a rate of ~15K/day. At 245K a sudden decrease in the intensity of elastic Rayleigh scattering was observed. This corresponds to an increase in the mean atomic vibration amplitude \( \sqrt{x^2} \) from ~0.15A to ~0.32A. After keeping the samples for ten hours near 245K the initial value of \( \sqrt{x^2} \) was recovered. The X-ray pattern of the sample show that this change in \( \sqrt{x^2} \) is connected with a structural re-arrangement. In the same temperature range thick samples undergo crystallisation where anomalies in \( \sqrt{x^2} \) are only weakly present.

Rayleigh scattering of Mössbauer radiation has proven to be a sensitive tool for studying lattice dynamical properties both in crystalline and non-crystalline materials. The basic theoretical and experimental outlines are described by O’Connor /1/. This method offers the important advantage that the scatterer must not necessarily contain a Mössbauer isotope. From the dependence of the relative intensities of the elastic and the inelastic scattering on sample temperature and scattering angle one obtains detailed information on the Debye-Waller factor; from the width of the scattered resonance line one deduces diffusional properties.

In non-crystalline materials like supercooled organic liquids the interest has concentrated on tracing the softening of these glasses from a decrease of the Debye-Waller factor in the regime some ten Kelvin above the glass transition point /2/. The non-equilibrium thermodynamic properties in the non-crystalline state are still not very well known in detail. A prominent experimental handicap is the poor reproducibility of data caused by minor differences in the thermal history of otherwise identical samples. For example, it has been shown recently /3/ that organic glasses doped with molecules containing iron as a Mössbauer probe, show drastic anomalies in the recoilfree fraction depending on different thermal treatments of the samples. These phenomena are caused by structural relaxation processes between different metastable states near the glass transition. The present experiments investigate the structural re-arrangement in amorphous semi-conductors which do not contain additional impurities of Mössbauer resonant atoms.

Samples of amorphous Te have been prepared by quenching the material from a molecular beam onto a 80K cold mylar substrate. The speed of deposition was typically ~5nm/s. The vacuum in the cryostat was kept \( \leq 10^{-6} \) torr. The experimental set-up for Rayleigh scattering is similar to the one described in /2/. As gamma ray source we used 100mCi of \(^{57}\)Co in Rh. The typical counting rate for a scattering angle of 29°30' and an acceptance angle of ~28' was about 3cts/s. Background corrections were carried out on the basis of countrates measured both off resonance and in resonance using an absorber of stainless steel (~54% absorption) first positioned between the source and scatterer and then switched to a position between scatterer and detector. In addition a small correction was included accounting for the background for resonant Rayleigh scattering from the sample holder and the substrate. The Debye-Waller factor was then obtained from the ratio of background corrected resonantly to total scattered...
intensity. The influence of Bragg peaks can be neglected due to the wide angle of acceptance in our scattering geometry. One may then simply set
\[ f_e \approx Q^2 \langle x^2 \rangle \]
with \( Q \) being the size of the scattering vector.

We found the root mean square displacement of Te atoms at 80K for thin amorphous samples \((<10^{5})\) to be \( \sqrt{x^2} \approx 0.08\AA \). The sample temperature was then increased in small steps with an average heating rate of ~15K/day. A smooth increase of \( \sqrt{x^2} \) to ~0.15\AA\ at 230K was seen. When reaching 245K a drastic increase of the inelastic together with a decrease of the elastic intensity occurs. The value of \( \sqrt{x^2} \) reaches ~0.32\AA. After about ten hours the original value of the rms displacement (~0.15\AA) is recovered. X-ray diffraction patterns taken in situ show that now the initially broad liquid-like peaks have sharpened. However, complete transformation to the usual trigonal crystalline phase did not take place. Further gradual heating to 320K followed by cooling to 80K does not result in further structural changes. Crystallization can only be achieved by a rapid temperature increase from 80K to room temperatures.

Thick samples, even when freshly prepared, show X-ray diffraction patterns at low temperatures similar to those found in thin samples after the annealing step near 250K. Between 250 and 260K a gradual crystallization is observed in thick samples. This process takes place without large changes in the Debye-Waller factor.

From electron diffraction data \(/4/\) it is known that the non-crystalline structure of vapour-quenched Te consists of short, distorted chains each containing only few molecular units. The present data show, that a defined structural change is present already below the crystallization point (~300K in case of fast annealing). We assume that in the temperature range of about 245K the slowly annealed samples have attained a metastable structure which undergoes stabilization on the time scale of several hours.

Due to the short chains the quenched sample contains originally a relatively large number of molecules situated at the end of chains. For these molecules additional degrees of freedom of motion are activated at the annealing point. This leads to a fast re-arrangement and to noticeable growth in length of chains. The rms displacement reaches at this temperature a value which corresponds to about 1/10 of the nearest neighbour separation and is thus comparable to the values found near the melting point. Final crystallization is accomplished by a gradual coalescence of the longer chains into the threefold spiral chains characteristic for crystalline Te. This process is obviously connected with far less dramatic local re-arrangement.

References


\(/4/\) Ichikawa, T., Phys. Status Solidi (b) 56 (1973) 707.