Low Temperature Vibrating Reed Measurements of Zr Based Metallic Glasses

C. Adler,[1] W. Mohr,[1] T. Peichl,[1] and G. Weiss[1, *]

[1]*Physikalisches Institut, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany*
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Vibrating reed measurements of the superconducting metallic glasses $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ and $Zr_{65}Al_{7.5}Cu_{27.5}$ are carried out in order to illuminate the influence of conduction electrons on the density of states of atomic tunneling systems, which dominate the low-temperature thermal and acoustic properties of disordered solids. The overall behavior of the two glasses is very similar and consistent with earlier experiments underlining the notion that the standard assumptions of the tunneling model are insufficient in the case of disordered metals. A quantitative comparison of the results reveal that the density of states of tunneling systems in the splat-cooled alloy $Zr_{65}Al_{7.5}Cu_{27.5}$ is about twice as high as in the bulk metallic glass $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$.

I. INTRODUCTION

The low-temperature thermal and acoustic behavior of glasses, dielectric or metallic, is dominated by two-level systems, which can be explained by the tunneling of small groups of atoms between two almost equivalent sites. Specific distributions of the relevant parameters are suggested by the well established tunneling model [1].

Acoustic measurements of various superconducting metallic glasses demonstrate that conduction electrons not only drastically change the dynamics of tunneling systems compared to insulating glasses where only phonons are important but also seem to influence the apparent density of states of the tunneling systems [2, 3]. These early experiments suffered from the fact that metallic glasses were only available as thin splats quenched from the liquid phase. The advent of bulk metallic glasses (BMG) considerably facilitated acoustic experiments and ultrasonic measurements in the 50 MHz range on the glassy alloy $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ essentially confirmed the older results [4].

II. EXPERIMENTS AND DISCUSSION

In order to quantify rigorously the influence of conduction electrons on the dynamics and finally on the density of states of the tunneling systems, measurements on the same sample are desirable over a wide frequency range. Our experiments on the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ aim in this direction. Here, we present first results of vibrating reed measurements at a frequency of 1.1 kHz [5] together with measurements of a splat-cooled $Zr_{65}Al_{7.5}Cu_{27.5}$ at a frequency of 2.98 kHz [6]. An important advantage of these alloys is
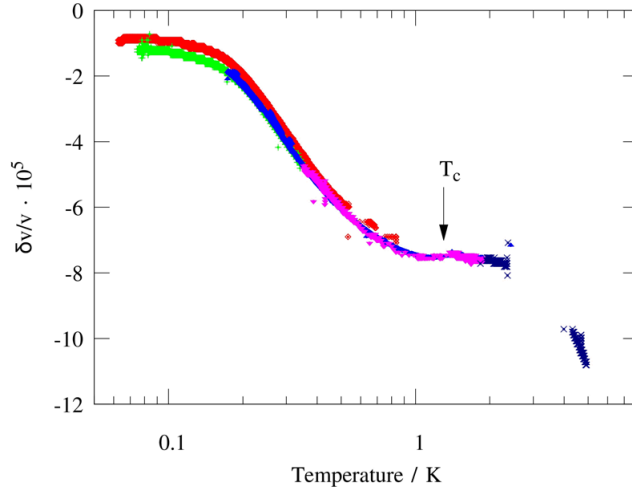
FIG. 1: Relative changes of sound velocity of the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ at low temperatures, compiled from different measurements. Respective data are identically normalized and thus illustrate the excellent repetitive accuracy across different runs. The arrow marks the superconducting transition temperature.

that they become superconducting below about 1.3 K and 1.7 K, respectively, which allows one to separate electron and phonon effects on the dynamics of the tunneling systems.

In our experiments, we measured relative changes of sound velocity, deduced from the reed's resonant frequency, and internal friction using special sample shapes to minimize uncontrollable losses of the clamping. During temperature changes, resonant frequency and amplitude were tracked by a computer-controlled phase-locked loop in combination with a digital lock-in amplifier and a frequency synthesizer. At various fixed temperatures, we determined absolute values of the internal friction by sweeping the driving frequency across the resonance. These data are marked by the large open symbols in our figures (see below).

Figure 1 presents the relative changes of the sound velocity with temperature of the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$. At very low temperatures, due to resonant interaction, the tunneling model predicts the sound velocity to increase with temperature [7]. Obviously, this is below the temperature range of our experiment. However, a short rise and a shallow maximum around 75 mK is noticeable. It is followed by a slow decrease, which in terms of the tunneling model, is due to relaxation processes where the tunneling systems relax via interaction with thermal phonons. Above 200 mK, the sound velocity decreases more steeply and then levels off around 1 K. At the superconducting transition $T_c \approx 1.3$ K (as determined by a susceptibility measurement), we observe a little cusp before the sound velocity decreases rapidly due to multi-phonon processes.

Relaxation of the tunneling systems via interaction with electrons additional to phonons should lead to a faster reduction of the sound velocity above $\approx T_c/2$. However, in contrast to our new and also previous [2, 3] measurements it is predicted that the steepest slope is right below T_c and a kink to a considerably smaller or even positive slope should
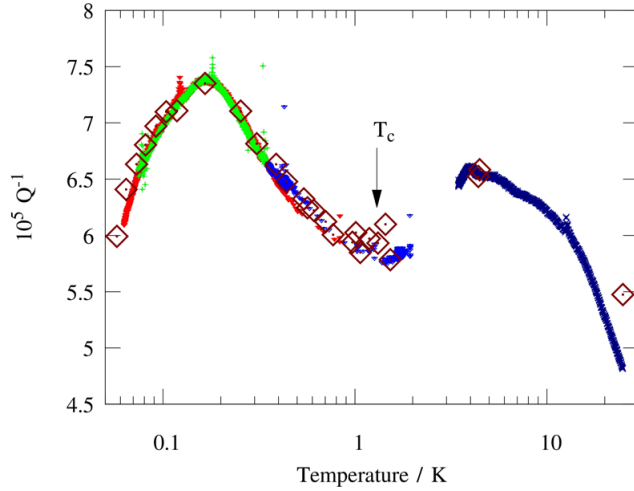
FIG. 2: Internal friction versus temperature of the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$. Data stem from frequency sweeps at fixed temperatures and evaluation of the resonance curves of the reed (large diamonds) or from tracking changes of the amplitude at the resonance frequency during slow temperature variations.

occur at T_c due to the rapidly closing energy gap at the transition from the superconducting to the normal conducting state [2, 8]. The discrepancy between this prediction and the experiment is not yet fully understood. Kagan and Prokof'ev [9] offered a possible explanation by introducing a modified density of states of the tunneling systems and including renormalization effects due to the interaction with electrons. However, to date, their idea has not been evaluated numerically far enough to prove its correctness.

Figure 2 shows the internal friction of the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ at low temperatures. In general, the behavior of the internal friction is consistent with earlier experiments [2, 3], and in fact, it is quite similar to our previous measurements [6] of another multi-component Zr-based, however, splat-cooled, glassy metal $Zr_{65}Al_{7.5}Cu_{27.5}$ (see figure 3).

From the lowest measured temperatures to 200 mK, internal friction increases and relaxation of the tunneling systems is caused by one-phonon processes. The subsequent maximum can be interpreted as the beginning of a plateau which is predicted by the tunneling model and found in experiments on dielectric glasses [7]. It is explained by the broad distribution of the relevant model parameters that characterize the two-level atomic tunneling systems. We like to note that in this temperature range the internal friction and the slope of the relative variation of the sound velocity agree quantitatively yielding a numerical value for the material parameter $C \approx 10^{-4}$ where C is given by Q^{-1} in the plateau range as $Q^{-1} = C\pi/2$. C contains the density of states of the tunneling systems and their squared deformation potential. In dielectric glasses C is usually larger by about a factor of 6.

The obvious deviation from a plateau found in our measurement demonstrates severe shortcomings of the standard model. The strong reduction of the internal friction above 200 mK coincides with the rapid decrease of the sound velocity. It marks the temperature
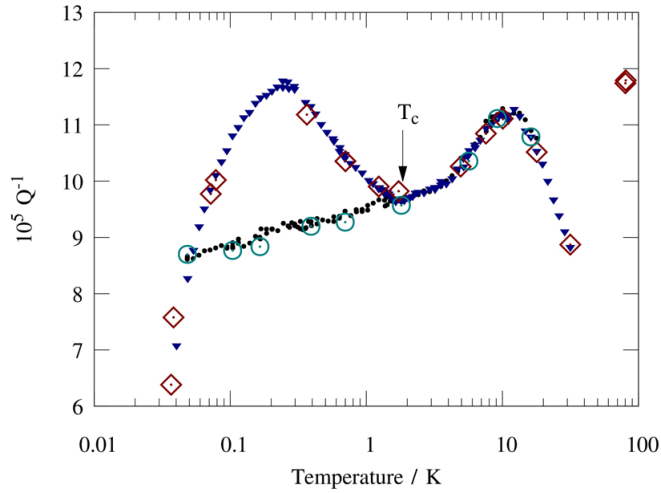
FIG. 3: Internal friction versus temperature of the metallic glass $Zr_{65}Al_{7.5}Cu_{27.5}$. The small triangles and open diamonds show measurements without magnetic field. The sample is superconducting below 1.7 K. Data shown as small dots and open circles were taken in a magnetic field of 4 T in order to suppress superconductivity.

where thermally excited quasi-particles start to play a role for the dynamics of the tunneling systems but also for their effective density of states. Again, the modifications suggested by Kagan and Prokof'ev [9], offer a semi-quantitative explanation.

Above T_c, around 4 K, the internal friction has another maximum. As in other glasses, this is caused by tunneling systems relaxing via multi-phonon-processes or thermal activation. There are, however, remarkable differences: Compared to other glasses, this maximum is much smaller and occurs at considerably lower temperatures. This can only be explained by limiting the density of states of the tunneling systems and, the limit has to be at much lower energies in the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ than in other, particularly dielectric glasses.

In figure 3, we present the temperature dependence of the internal friction of the splat-cooled metallic glass $Zr_{65}Al_{7.5}Cu_{27.5}$. Its behavior in the superconducting state and in the normal conducting state at temperatures above $T_c \approx 1.7$ K resembles closely that of our BMG sample. Quantitatively, we observe some differences. The internal friction of $Zr_{65}Al_{7.5}Cu_{27.5}$ is overall considerably higher by factor 1.5 to 2 and it drops off at higher temperature compared to the BMG. Both these observations might be related to the largely different cooling rates during solidification of the two chemically similar glasses and imply a higher density of states and higher cut-off energy of atomic tunneling systems in the more rapidly solidified glass. In both samples we find a minimum of the internal friction around 50 K followed by a steep increase towards the glass transition. At room temperature internal friction takes values of the order 10^{-3}.

Figure 3 additionally shows measurements of $Zr_{65}Al_{7.5}Cu_{27.5}$ in a magnetic field of 4 T, sufficiently high to suppress superconductivity. We see that in the normal conduct-

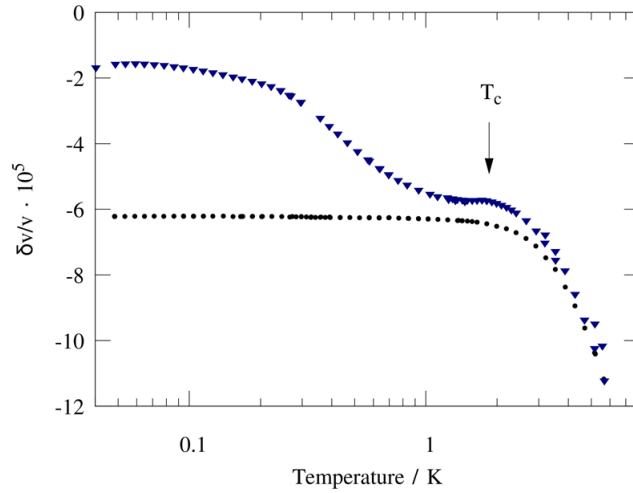FIG. 4: Relative change of the sound velocity of the splat-cooled metallic glass $Zr_{65}Al_{7.5}Cu_{27.5}$. The triangles show measurements without magnetic field, i.e. the sample is superconducting below 1.7 K. The data shown as small dots were taken in a magnetic field of 4 T in order to suppress superconductivity.

ing state the internal friction gradually and monotonically decreases on cooling down to the lowest temperatures of our experiment. Around 100 mK Q^{-1} in the normal state is about 25% smaller than in the superconducting state. The behavior is similar to previous experiments [2, 3] and evidence for the inadequacy of the standard tunneling model to describe metallic glasses. With standard assumptions for the distribution of parameters the tunneling model predicts that the internal friction is independent of temperature in this temperature range and its value is independent of the sample being superconducting or normal conducting [7].

Finally, in figure 4 we plotted the temperature dependence of the sound velocity of $Zr_{65}Al_{7.5}Cu_{27.5}$ both with and without magnetic field. The behavior in the superconducting state is similar to our BMG sample. The expected shallow maximum around 60 mK is clearly visible and the subsequent $\log T$ decrease due to relaxation processes via thermal phonons extends to higher temperatures. The more rapid decrease above 300 mK is less pronounced than in the BMG, however, we again observe a cusp around T_c which is a severe contradiction to the standard theory as stated already above. When the sample is held normal conducting in a magnetic field, the sound velocity is almost temperature-independent between 50 mK and 1.5 K. In terms of the tunneling model, this would require an extremely delicate and thus unexpected balance between resonant and relaxation processes.

III. CONCLUSIONS

In summary, the measured temperature dependencies of both, internal friction and sound velocity of the BMG $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ and of the splat-cooled metallic glass $Zr_{65}Al_{7.5}Cu_{27.5}$ confirm the deviations from the tunneling model found by previous measurements of other superconducting metallic glasses. The deviations might be explained by the influence of conducting electrons, which not only drastically change the dynamics of tunneling systems but also seem to influence the apparent density of states of tunneling systems.

Acknowledgments

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 * Electronic address: georg.weiss@kit.edu

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