Coherent echoes in crystals with tunneling systems in magnetic fields

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Abstract

At low temperatures the acoustic and dielectric properties of disordered solids are strongly influenced by the presence of atomic tunneling systems. The recent discovery of unexpected magnetic field effects in the dielectric properties of non-magnetic glasses has raised the question whether, and how, atomic tunneling systems can couple to external magnetic fields. We have investigated well-defined tunneling systems in alkali halide crystals, such as the mixed crystal $(KBr)_{1-x}(KCN)_x$, by means of coherent polarization echoes. We find a very strong non-monotonic dependence of the amplitude of spontaneous echoes in these crystals on the applied magnetic field, indicating a direct coupling of the magnetic field to atomic tunneling systems.

Key words: disordered solids; tunneling systems; coherent properties;

Until very recently it was the general believe that the dielectric properties of insulating glasses – free of magnetic impurities – are largely independent of external magnetic fields. However, new investigations have shown that the low-temperature dielectric properties of certain multi-component glasses are extremely sensitive to magnetic fields. In particular, the low-frequency dielectric susceptibility [1–4] and the amplitude of spontaneous polarization echoes [5] show a striking non-monotonic dependence on the applied magnetic field.

The low-temperature dielectric properties of glasses are governed by atomic tunneling systems, which arise from the amorphous structure. Therefore, it seems natural to assume that the tunneling systems are responsible for the magnetic field dependence. In fact, two models have been suggested to explain the magnetic field effect, which are based on the assumption of a direct coupling of magnetic fields to tunneling systems [6,7]. A verification of theses theoretical models, however, suffers from the fact that the microscopic nature of the tunneling systems in amorphous solids is hitherto unknown. Therefore we have studied two crystalline defect systems, whose low-temperature properties are also strongly influenced by the presence of tunneling states. In these cases, however, a much better understanding of the microscopic origin of the tunneling systems is available. We have investigated these samples be means of coherent polarization echoes. This technique has the advantage, that exclusively tunneling states contribute to the signal and therefore any magnetic field dependence observed with this technique has to be related to the properties of the tunneling systems. In addition, polarization echoes are a resonant experiment, which means that only tunneling systems in resonance with the exciting microwave field are observed.

The first crystalline defect system investigated in these experiments is $(KBr)_{1-x}(KCN)_x$. This mixed crystal, is often referred to as a model system for the tunneling states in glasses, because in a certain concentration range it exhibits far reaching similarities of its low-temperature properties with those of amorphous materials (e.g. [8]). At concentrations $x < 0.1$, the systems that contribute to the echo experiments are well-defined pairs, consisting of CN^- ions on next nearest neighbor sites [9]. Fig. 1 shows the amplitude of two pulse echoes in a $(KBr)_{0.925}(KCN)_{0.075}$ crystal

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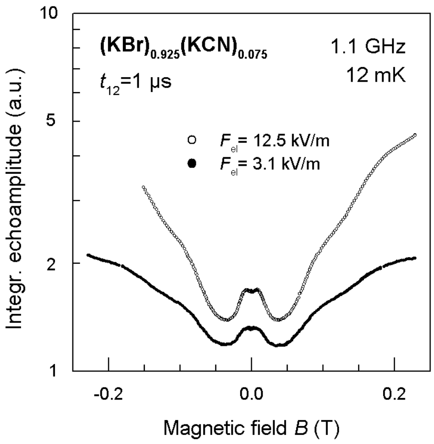

Fig. 1. Magnetic field dependence of the amplitude of two-pulse echoes generated in $(KBr)_{0.925}(KCN)_{0.075}$ excited at two different electric field strengths.

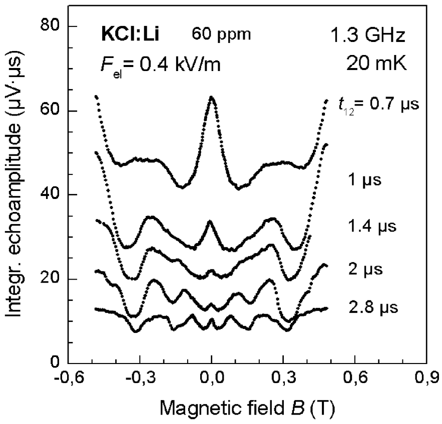

Fig. 2. Magnetic field dependence of the amplitude of two-pulse echoes generated in KCl containing 60 ppm of Li for different delay times t_{12} between the exciting pulses.

as a function of the applied magnetic field and for two different electrical field strengths of the exciting pulses. Clearly, a strong influence of the magnetic field on the echo amplitude is observed. The overall pattern is similar to that previously observed for the multicomponent glass a-BaO-$Al_2O_3SiO_2$ [5]. The dependence on the amplitude of the exciting pulses suggests that the magnetic field effect is non-linear with respect to the magnitude of the electrical field strength.

The other system that we have investigated is KCl containing small amounts of Li, that substitutes for potassium forming off-center tunneling states. At very low concentrations, the mean interaction between the Li defects is negligible and the majority of tunneling states are symmetric and identical. In this case the system can be understood on a microscopic basis and the corresponding parameters are well known [10]. However, as in the case of CN^- in KBr, the echo is caused by pairs of strongly coupled Li ions, which occur statistically – in a small number – even at very low Li concentration [11]. Fig. 2 shows the amplitude of polarization echoes generated in a KCl crystal containing 60 ppm of lithium as a function of the applied magnetic field for different delay times t_{12} between the two exciting pulses. Again we observe a striking non-monoton magnetic field dependence, indicating that the magnetic field effect is a general phenomenon of tunneling states and not just a peculiarity of a certain glass. Interestingly, the pattern of the magnetic field effect, depends strongly on the delay time, showing that the free development of the phase of the tunneling systems is affected by the magnetic field. The fact, that we observe a magnetic field dependence of the echo amplitude in this sample clearly rules out collective effects as an origin for these phenomena as suggested by [6], because at 60 ppm the number density of pairs seems much to small for a collective behavior. Therefore, we conclude that the magnetic field effect must be due to individual tunneling systems. Note that, the pairs of coupled Li ions in KCl can be treated as individual tunneling systems, because the coupling is so strong that only a common, fully coherent tunneling motion of both Li ions is possible. Therefore it seems that also the model proposed by Würger [7] can be ruled out as a theoretical basis to explain our data, because in this model pairs of weakly interacting tunneling systems are considered.

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