Anisotropic glass-like properties in tetragonal disordered crystals

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Abstract. The low temperature acoustic and thermal properties of amorphous, glassy materials are remarkably similar. All these properties are described theoretically with reasonable quantitative accuracy by assuming that the amorphous solid contains dynamical defects that can be described at low temperatures as an ensemble of two-level systems (TLS), but the deep nature of these TLSs is not clarified yet. Moreover, glassy properties were found also in disordered crystals, quasicrystals, and even perfect crystals with a large number of atoms in the unit cell. In crystals, the glassy properties are not universal, like in amorphous materials, and also exhibit anisotropy. Recently it was proposed a model for the interaction of two-level systems with arbitrary strain fields (Phys. Rev. B 75, 64202, 2007), which was used to calculate the thermal properties of nanoscopic membranes at low temperatures. The model is also suitable for the description of anisotropic crystals. We describe here the results of the calculation of anisotropic glass-like properties in crystals of various lattice symmetries, emphasizing the tetragonal symmetry.

1. Introduction

The low temperature acoustic and thermal properties of amorphous, glassy materials are remarkably similar and they can be explained—to a large extent—by assuming that the material contains a large number of dynamic defects. These dynamic defects are tunneling systems (TS) which are modeled by an ensemble of two-level systems (TLS)\textsuperscript{1,2}. Crystals with defects—with a large enough amount of disorder—exhibit also glass-like properties, but these properties are not so universal and, even more, they are not isotropic—for example the absorption and the propagation velocity change of elastic waves depend on the propagation and polarization directions of the wave\textsuperscript{3}.

A microscopic model of the tunneling systems is still missing. For this reason, the study of disordered crystals is especially interesting because it offers additional information: in some materials we know quite well which are the entities that tunnel between different equilibrium positions. Beside this, the anisotropy of the TLS-sound wave interaction in crystals represents another challenge to the interaction models of TLSs which requires clarification.

A free TLS is described in a two-dimensional Hilbert space by the Hamiltonian

\[ H_{\text{TLS}} = \frac{\Delta}{2} \sigma_z + \frac{\Lambda}{2} \sigma_x \equiv \frac{1}{2} \begin{pmatrix} \Delta & \Lambda \\ -\Lambda & \Delta \end{pmatrix} \]  

(1)
where \( \sigma_z \) and \( \sigma_x \) are Pauli matrices; the parameters \( \Delta \) and \( \Lambda \) are called the \textit{asymmetry of the potential} and \textit{tunnel splitting}, respectively. The excitation rate of the TLS, \( \epsilon \), is obtained by diagonalizing \( H_{\text{TLS}} \) and this gives \( \epsilon = \sqrt{\Delta^2 + \Lambda^2} \).

The perturbation of the free TLS, induced by a strain field is \( H_{\text{I}} \equiv (\delta/2)\sigma_z \), where in general \( \delta \equiv 2\gamma_{ij}S_{ij} \), with \( S_{ij} \) being the strain tensor and \( \gamma_{ij} \) a symmetric tensor that characterizes the TLS and its interaction with the strain field—here, as everywhere in this paper, we assume summation over repeated indices. In bulk isotropic materials, like in amorphous solids, the elastic waves may be decomposed into longitudinally and transversely plane waves and the interaction term \( \delta \) may be written as

\[
\delta = 2\gamma_{ij}S_{ij},
\]

where \( \sigma \) is the polarization, \( t \) (longitudinal) or \( t \) (transversal), of the perturbing wave. In this case the absorption rate of a phonon of polarization \( \sigma \) and wavevector \( k \), by a TLS is

\[
\Gamma_{k,\sigma} = \frac{2\pi}{\hbar} \gamma_{\sigma}^2 \frac{\hbar k}{2V\rho c_{\sigma}} \frac{\Lambda^2}{\epsilon^2} n_{k,\sigma} \delta(hc_\sigma k - \epsilon),
\]

where by \( n_{k,\sigma}, \rho, c_{\sigma} \), and \( V \) we denote the population of the phonon mode, the density of the solid, the sound velocity and the volume of the solid, respectively. If the number of TLSs of asymmetry and tunnel splitting in the intervals \( d\Delta \) and \( d\Lambda \) is \( P(\Delta, \Lambda)d\Delta d\Lambda = V\rho/\Delta d\Delta d\Lambda \), then integrating \( \Gamma_{k,\sigma} \) over the TLS ensemble and using \( n_{k,\sigma} = \exp(\beta hc_\sigma k) - 1 \^{-1} \), we obtain the \textit{standard} total phonon absorption rate,

\[
\left(\epsilon^{(S)}_{k,\sigma}\right)^{-1} = P_k \gamma_{\sigma}^2 \kappa \cdot \tanh \left(\frac{\epsilon}{2k_B T}\right),
\]

where we introduced the short-hand notation \( K \equiv \pi n_{k,\sigma} k/(V\rho c_{\sigma}) \).

Although widely used, the simplified version of the tunneling model exposed above cannot account for the anisotropy of the glass-like properties observed in disordered crystals. To explain this anisotropy, in Refs. \[4,5\] we applied the model introduced in \[6\]. In this model, a direction \( \mathbf{t} \) is associated to each TLS \( \mathbf{t} \equiv (t_x, t_y, t_z)^t \), where the superscript \( t \) denotes the transpose of a matrix or vector and \( \delta \) was written in the form \[6,7,8\]

\[
\delta = 2\mathbf{T}^t \cdot [\mathbf{R}] \cdot \mathbf{S},
\]

with \( \mathbf{T} \equiv (t_x^2, t_y^2, t_z^2, 2t_y t_z, 2t_z t_x, 2t_x t_y)^t \) and \( \mathbf{S} \) is the strain tensor in abbreviated subscript notations. The \( 6 \times 6 \) symmetric matrix \([\mathbf{R}]\) contains the TLS-strain field coupling constants \( r_{ij} \) and its structure is determined by the symmetries of the lattice in which the TLS is embedded. In Refs. \[4,5\] we applied this model to calculate the glassy properties of disordered cubic (Ca stabilized zirconium \[9\]), trigonal (neutron irradiated quartz \[10\]) and hexagonal crystals. Here we shall report results on tetragonal lattices.

2. Phonon scattering rates in tetragonal lattices

In the new model, the phonon absorption rate by a TLS is

\[
\Gamma_{k,\sigma}(\mathbf{t}) = \frac{2\pi}{\hbar} \frac{\Lambda^2 n_{k,\sigma}}{\epsilon^2} |\mathbf{T}^t \cdot [\mathbf{R}] \cdot \mathbf{S}_{k,\sigma}|^2 \delta(\epsilon - \hbar\omega).
\]

We see that the main characteristic of the TLS-phonon interaction is contained in the quantity \( \mathbf{M}_{k,\sigma}(\mathbf{t}) \equiv \mathbf{T}^t \cdot [\mathbf{R}] \cdot \mathbf{S}_{k,\sigma} \), which bears an intrinsic anisotropy through the
matrix \([R]\), on which the symmetries of the lattice are imposed \([11; 12; 13]\). To calculate the average scattering rate of a phonon by the ensemble of TLSs, now we have to average not only over the distribution of \(\Delta\) and \(\Lambda\), but also over the distribution of \(t\). In this way we get the total phonon absorption rate,

\[
\tau_{k\sigma}^{-1} = \frac{2\pi P_0 \tanh \left( \frac{2\pi \sigma T}{\hbar} \right)}{n_{k\sigma} |M_{k\sigma}(\hat{t})|^2}. \tag{7}
\]

To reduce the number of degrees of freedom of the problem, in what follows we shall assume that \(\hat{t}\) is isotropically oriented.

For a tetragonal lattice of symmetry classes \(4mm, 422, \overline{4}2m, 4/mmm\), the matrix \([R]\) has the form \([11]\)

\[
[R] = \begin{pmatrix}
  r_{11} & r_{12} & r_{13} & 0 & 0 & 0 \\
  r_{12} & r_{11} & r_{13} & 0 & 0 & 0 \\
  r_{13} & r_{13} & r_{33} & 0 & 0 & 0 \\
  0 & 0 & 0 & r_{44} & 0 & 0 \\
  0 & 0 & 0 & 0 & r_{44} & 0 \\
  0 & 0 & 0 & 0 & 0 & r_{66}
\end{pmatrix}, \tag{8}
\]

similar to that of the tensor of elastic stiffness constants, \([c]\). The difference between the trigonal lattice of symmetry \(32\) and the chosen classes of tetragonal lattice is that \(r_{14}\) and \(c_{14}\) are zero. In that sense the hexagonal and tetragonal lattices are similar to each other. However, in contrast to both hexagonal and trigonal \(32\) systems, for the tetragonal lattice the condition \(r_{66} = (r_{11} - r_{12})/2\) does not hold anymore. As a consequence, the number of independent constants is equal to that for trigonal \(32\) lattice, whereas the structure of \([R]\) is similar to that for the hexagonal lattice.

For concreteness, we shall consider the class \(422\), with a system of coordinates chosen such that the \(z\) and \(x\) axes are the 4-fold and 2-fold rotational symmetry axes, respectively, while the \(y\) axis is perpendicular to both \(x\) and \(z\). Solving the Christoffel equation we find that the crystal can sustain pure transversal waves in all three directions, \(x\), \(y\), and \(z\). For the transversal waves propagating in the \(x\) direction, the ones polarized in the \(z\) direction propagate with the velocity \(\sqrt{c_{44}/\rho}\) whereas the ones polarized in the \(y\) direction propagate with the velocity \(\sqrt{c_{66}/\rho}\). The transversal waves propagating in the \(y\) direction are similar to the ones propagating in the \(x\) direction: the waves polarized in the \(x\) direction have a sound velocity of \(\sqrt{c_{44}/\rho}\), whereas the ones polarized in the \(z\) direction have a sound velocity of \(\sqrt{c_{44}/\rho}\). Finally, the transversal waves propagating in the \(z\) direction have all the same sound velocity, \(\sqrt{c_{44}/\rho}\).

Averaging over the directions \(\hat{t}\) we obtain for the transversely polarized waves propagating in the \(x, y\) and \(z\) directions the results

\[
\langle |M_{k\hat{x},t}|^2 \rangle = \langle |M_{k\hat{y},t}|^2 \rangle = \frac{4N^2k^2r_{66}^2}{15}, \tag{9a}
\]

and

\[
\langle |M_{k\hat{z},t}|^2 \rangle = \langle |M_{k\hat{y},t}|^2 \rangle = \langle |M_{k\hat{x},t}|^2 \rangle = \langle |M_{k\hat{x},t}|^2 \rangle = \frac{4N^2k^2r_{44}^2}{15}. \tag{9b}
\]

in obvious notations: the first subscript indicates the propagation direction and the second denotes the direction of polarization. Plugging equations \((9a)\) and \((9b)\) into \((7)\) we obtain the general type of expression

\[
\left( \frac{\tau_{k\sigma}^{-1}(T)}{\rho} \right)^{-1} = P_0 \langle \gamma_{k\sigma}^{(T)} \rangle^2 K \cdot \tanh \left( \frac{\epsilon}{2k_B T} \right), \tag{10}
\]

where \(\epsilon\) is the phonon energy.
where the superscript \(^{(T)}\) comes from the tetragonal symmetry. Concretely, the new \(\gamma\) constants are

\[
(\gamma^{(T)}_{k\hat{y},\hat{x},t})^2 = (\gamma^{(T)}_{k\hat{x},\hat{y},t})^2 = \frac{4r_{66}^2}{15}, \\
(\gamma^{(T)}_{k\hat{x},\hat{z},t})^2 = (\gamma^{(T)}_{k\hat{z},\hat{y},t})^2 = (\gamma^{(T)}_{k\hat{y},\hat{z},t})^2 = \frac{4r_{44}^2}{15}.
\]  

(11a)

(11b)

The ratio \(r_{66}/r_{44}\) can be obtained by measuring the characteristics (sound velocity change or attenuation rate) of elastic waves propagating along the \(\langle 100 \rangle\) crystallographic directions. It is interesting to note now the similarity between relations like

\[
\frac{\tau^{(T)}_{k\hat{x},\hat{z},t}}{\tau^{(T)}_{k\hat{x},\hat{y},t}} = \left(\frac{\gamma^{(T)}_{k\hat{x},\hat{y},t}}{\gamma^{(T)}_{k\hat{x},\hat{z},t}}\right)^2 = \left(\frac{r_{66}}{r_{44}}\right)^2
\]

and relations like

\[
\left(c^{(T)}_{k\hat{x},\hat{z},t}/c_{k\hat{x},\hat{y},t}\right)^2 = \frac{c_{66}}{c_{44}}
\]

that hold for tetragonal lattices—e.g. \(c_{k\hat{x},\hat{y},t}\) is the velocity of a transversal sound wave propagating along the \(x\) direction and polarized along the \(y\) direction.

3. Conclusions

We calculated the average scattering rates of pure transversal phonon modes on the TLSs in tetragonal disordered crystals, to emphasize the anisotropy of the glass-like properties imposed by the lattice anisotropy. Two of the parameters of the model—the TLS-strain field coupling constants—may be obtained by measuring attenuation rates or sound velocity changes of elastic waves propagating along the different \(\langle 100 \rangle\) directions. Determining the coupling constants enables one to calculate the glassy properties of the crystal in any direction.

The calculations can be extended easily to disordered crystals of any symmetry. Moreover, although we used in our calculations an isotropic distribution over the TLS orientations, the comparison of our calculations with experimental data would enable one to find if our assumption is true or not. If it is not true, one can determine, at least in principle, the distribution of the orientations of the TLSs.

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