ANISOTROPIC INTERACTION BETWEEN TWO-LEVEL SYSTEMS

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Abstract. Using the model of interaction between two-level systems (TLS) and strain fields introduced in Phys. Rev. B 75, 064202 (2007), we calculate the TLS-TLS interaction. We particularize our calculations to amorphous materials and analyze the dependence of the interaction Hamiltonian on the orientations of the two TLSs and of the direction that connects them. Assuming isotropic distribution of the TLSs orientations, we then averaged the interaction over one of the TLSs orientations. The resulting interaction depends in a simple way on the angle between the orientation of the other TLS and the line that connects the two TLSs.

Key words:

1 INTRODUCTION

Low temperature thermal properties of dielectric crystals differ markedly from those of dielectric amorphous solids. In crystals at low temperatures, the thermal properties are determined mainly by the acoustic phonons and their interaction with the defects; the system is well described by the Debye model and this renders the well known \( T^3 \) dependence of the specific heat (by \( T \) we denote the temperature) in a temperature range below 10 K. The heat conductivity of crystals is strongly dependent on the chemical composition of the crystal and on the nature and density of lattice defects.

In contrast to the crystals, the specific heat of amorphous, glassy materials below 1 K is proportional to the temperature, whereas the heat conductivity varies approximately like \( T^2 \). Moreover-and this is maybe the most interesting characteristic of glassy materials-, some of their thermal properties (e.g. the heat conductivity) are (quasi)universal, i.e. they have a very weak dependence on the chemical composition or disorder of the solid.

The thermal properties of glassy materials are described mainly in what is now called the standard tunneling model (STM) [1, 2] and have been attributed to the existence of dynamical defects in the solid. These dynamical defects are atoms or groups of atoms which tunnel between the two potential minima of quasi-symmetric two-well potentials. Because of the quasi-symmetry of the
potential landscape, the two lowest energy levels of the system may be very close to each-other and therefore may have a significant contribution to the low temperature properties of the solid. If the potential barrier between the two wells is high enough, then the energy separation between the second and the third energy levels is much bigger than the energy separation between the lowest two energy levels and the system is well described by a two-level system (TLS), in a two-dimensional Hilbert space. In this Hilbert space, the Hamiltonian of a single TLS may be written as

\[ H_{\text{TLS}} = \frac{1}{2} \left( \begin{array}{cc} \Delta & -\Lambda \\ -\Lambda & -\Delta \end{array} \right) \equiv \frac{\Delta}{2} \sigma_z - \frac{\Lambda}{2} \sigma_x, \] (1)

where \( \sigma_z \) and \( \sigma_x \) are Pauli matrices. The parameters \( \Delta \) and \( \Lambda \) in Eq. (1) are called the asymmetry of the potential and the tunneling splitting, respectively.

An elastic wave or a deformation of the solid perturbs the “free” TLS hamiltonian (1) by

\[ H_I = \frac{1}{2} \left( \begin{array}{cc} \delta & 0 \\ 0 & -\delta \end{array} \right), \] (2)

where in general we can write \( \delta \equiv 2\gamma_{ij}S_{ij} \), where \( S_{ij} \) is the strain tensor of the deformation field and \( \gamma_{ij} \) is a symmetric tensor that characterizes the TLS and its interaction with the deformation field–throughout this paper we assume summation over the repeated indices. While the tensor \( [\gamma] \) is assumed to be the same for all TLSs, the parameters \( \Delta \) and \( \Lambda \) vary form one TLS to the other, with a probability distribution

\[ P(\Delta, \Lambda) = \frac{V}{P_0/\Lambda}, \] (3)

where \( V \) is the volume of the solid and \( P_0 \) is a constant.

Although the STM describes well most of the properties of amorphous materials, it fails to explain certain “details”, or some of its features need deeper understanding. One of the details that STM cannot explain in its simplest form is the fact that the heat conductivity is not exactly proportional to \( T^2 \), but it is rather proportional to \( T^{1.8} \). Nevertheless, the most challenging problem related to amorphous materials is the explanation of the “universality” of their thermal properties. Why such a wide variety of materials (polymers, oxide glasses, polycrystalline metals, etc.) have such similar properties and why the probability distribution \( P(\Delta, \Lambda) \) has the simple form given by Eq. (3) for all of them? These questions are not yet answered, but there are hints that these effects are due to the interaction between the TLSs.
1.1 TLS-TLS INTERACTION IN THE STM

The total Hamiltonian of an amorphous solid may be split into three parts: the free phonons Hamiltonian, the defects (i.e. the TLSs) Hamiltonian, and the interaction Hamiltonian between phonons and TLSs:

\[ H = H_{ph} + H_{def} + H_{int}, \quad (4) \]

where

\[ H_{ph} = \sum_{k,\sigma} \left( \frac{|p_{k,\sigma}|^2}{2M} + M\omega_{k,\sigma}^2 \frac{|u_{k,\sigma}|^2}{2} \right) \quad (5a) \]

\[ H_{def} = \sum_{m} H_{TLS}(m) \quad (5b) \]

\[ H_{int} = -\sum_{m} [\gamma_{ij} S_{ij}(m)\sigma_z] \quad (5c) \]

In Eq. (5a), the summation is taken over all the phonon modes, with \( k \) denoting the phonon wavenumber and \( \sigma \), the phonon’s polarization; since we work with isotropic amorphous solids, the phonons will be considered as simple longitudinally or transversally polarised elastic waves (\( \sigma = l \) or \( t \)). Also in Eq. (5a), \( M \) is the mass of the elementary “cell” of the medium, whereas \( p_{k,\sigma} \) and \( u_{k,\sigma} \) are the momentum and displacement operators, respectively.

In equations (5b) and (5c), the summations are taken over the TLSs in the system, therefore \( H_{TLS}(m) \) is the free Hamiltonian (1) of the \( m \)-th TLS and \( S_{ij}(m) \) is the strain field produced by the phonon system at the \( m \)-th TLS.

The interaction of the TLSs with the strain fields changes the ground state of the system, in the sense that the lowest energy state is not the one of zero strain, but one of non-zero strain. This non-zero strain that appears in the solid body at equilibrium, with the TLSs frozen in a configuration \( s(m), \ m = 1, 2, \ldots \), induces long-range interaction between the TLSs. This interaction produces chaotic shifts of the energies of the TLSs, giving rise to spectral diffusion, which is the random change of the energy of a specific TLS due to the relaxation of the environment and is clearly manifested in low temperature hole-burning and phonon-echo experiments [3].

2 ANISOTROPIC TLS-TLS INTERACTION

Minimizing the total energy (4) of the system one can obtain the interaction energy between the TLSs,

\[ V_I = U_{12}\sigma_{1z}\sigma_{2z}. \quad (6) \]
where \( \sigma_{1z} \) and \( \sigma_{2z} \) are the Pauli matrices associated to the interacting TLSs, 1 and 2, respectively. The coupling energy, \( U_{12} \), in a three-dimensional bulk material, has the expression (see for example [4])

\[
U_{12} = -\frac{1}{2\pi R^3 \rho c_t^2} \sum_{ijkl} \gamma_{1ij} \gamma_{2ik} (\delta_{jk} - 3n_j n_k) \\
+ \frac{1}{2\pi R^3 \rho} \left( \frac{1}{c_l^2} - \frac{1}{c_t^2} \right) \sum_{ijkl} \gamma_{1ij} \gamma_{2kl} \left[\delta_{il} \delta_{jk} + \delta_{ij} \delta_{lk} \right] + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} + 3(\delta_{ij} n_k n_l + \delta_{ik} n_j n_l + \delta_{il} n_j n_k) + \delta_{jk} n_i n_l + \delta_{jl} n_i n_k + \delta_{il} n_j n_k - 15n_i n_j n_k n_l \right]
\]

where by \( \hat{n} \) we denote the direction from the TLS 1 to the TLS 2 and \( R \) is the distance between them; \( c_l \) and \( c_t \) are the longitudinal and transversal sound velocities, respectively, \( \rho \) is the mass density of the solid, and \( \gamma_{1,2ij} \) are the components of the tensors of coupling constants between the TLS 1 or 2 and a deformation strain field in the body. The second rank tensors will be denoted in general as \( \gamma_{ijkl} \). From (7) follows that \( U_{12} \) is proportional to \( R^{-3} \)-like in the dipol-dipol interaction—and can be written as \( U_{12} \equiv u_{12} / R^3 \). Moreover, one can easily show that the average of \( U_{12} \) over the direction \( \hat{n} \), denoted as \( \langle U_{12} \rangle \hat{n} \) is zero for any \( \gamma_{1,2ij} \). Based on this, and since the expression of \( U_{12} \) is very complicated for general tensor elements, \( \gamma_{1,2ij} \)—which are not known in detail anyway—, the usual assumption one does is that \( U_{12} \) is randomly distributed (i.e. there is no specific orientation dependence in the interaction) and a method similar to the random 3D Ising model is employed in analysing the system [4].

In Ref. [5] we introduced a model in which a direction, call it \( \hat{t} \), is associated to each TLS and the TLS-strain field interaction depends on the relative orientation of \( \hat{t} \) with respect to the strain field. From the components of \( \hat{t} \) we construct the symmetric second rank tensor \( [T] \), with \( T_{ij} = t_i t_j \) and we write the \( [\gamma] \) tensor in the general form, \( \gamma_{ij} = T_{kl} R_{ijkl} \). Then the forth rank tensor \( [R] \) is actually the tensor of coupling constants between the TLS and the deformation field and its general structure is determined by the symmetry properties of the solid in which the TLS is embedded. For an isotropic medium, \( [R] \) has only two independent parameters, \( \zeta \) and \( \xi \), similar to the Lamé constants that enter in the construction of the elastic stiffness constants in elasticity: \( R_{ijkl} = [\zeta' \delta_{ij} \delta_{kl} + \zeta (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})] \). Usually, for a direct connection to the STM, we take out a factor \( \tilde{\gamma} = \zeta' + 2\xi' \) and write \( R_{ijkl} = \tilde{\gamma} (\delta_{ij} \delta_{kl} + \xi (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})) \), where \( \zeta = \zeta' / \tilde{\gamma} \) and \( \xi = \xi' / \tilde{\gamma} \); this implies \( \zeta + 2\xi = 1 \) Using the expressions for \( R \) and \( T \), the \( [\gamma] \) tensors become \( \gamma_{1,2ij} = \tilde{\gamma} (\delta_{ij} + 2\xi t_{1,2j} t_{1,2j}) \). If we plug this into (7) and perform all the summations, we get the much
In Eqs. (8) and (9), we can observe from Eq. (10) that the average of \( \langle \hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}} \rangle \) gives zero, as one would expect. Moreover, the TLSs placed along the direct ion \( \hat{\mathbf{t}}_1 \) and get simpler expressions,

\[
U_{12} = \frac{\tilde{\gamma}^2}{2\pi R^3 \rho} \left\{ 6\xi(3\xi + 2\xi)c_i^{-2} - 2(\xi + \xi)c_i^{-2} \right\} [ (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 + (\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})^2 ]
+ 12\xi^2 [5c_i^{-2} - 4c_i^{-2}] (\hat{\mathbf{t}}_1 \cdot \hat{\mathbf{t}}_2) (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})(\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})
- 60\xi^2 (c_i^{-2} - c_i^{-2})(\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 (\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})^2 - 4\xi^2 (3c_i^{-2} - 2c_i^{-2})(\hat{\mathbf{t}}_1 \cdot \hat{\mathbf{t}}_2)^2
- 4\xi (\xi + 2\xi)(c_i^{-2} - c_i^{-2}) - 4\xi \xi c_i^{-2} \right\}
\tag{8}
\]

Using the relation \( \zeta + 2\xi = 1 \), we eliminate \( \zeta \) from all the expression above and get

\[
U_{12} = \frac{\tilde{\gamma}^2 \xi}{\pi R^3 \rho} \left\{ 3[(3 - 4\xi)c_i^{-2} - 2(1 - \xi)c_i^{-2}] [ (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 + (\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})^2 ]
+ 6\xi [5c_i^{-2} - 4c_i^{-2}] (\hat{\mathbf{t}}_1 \cdot \hat{\mathbf{t}}_2) (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})(\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})
- 30\xi (c_i^{-2} - c_i^{-2})(\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 (\hat{\mathbf{t}}_2 \cdot \mathbf{\hat{n}})^2 - 2(3\xi - 2)(\hat{\mathbf{t}}_1 \cdot \hat{\mathbf{t}}_2)^2
- 2(2 - 3\xi)(c_i^{-2} - c_i^{-2}) - 2(1 - 2\xi)c_i^{-2} \right\}
\tag{9}
\]

In Eqs. (8) and (9), \( U_{12} \) depends on the angles between \( \hat{\mathbf{t}}_1 \), \( \hat{\mathbf{t}}_2 \), and \( \mathbf{\hat{n}} \). The dependence is very complicated, but if we assume that in any volume element the TLSs are isotropically oriented, we can calculate the average interaction of TLS 1 with the TLSs located in a small volume \( \delta V \), at position \( \mathbf{\hat{n}}R \) from TLS 1, by averaging over \( \hat{\mathbf{t}}_2 \). This is simply done and we obtain

\[
\langle U_{12} \rangle_{\hat{\mathbf{t}}_2} = \frac{\tilde{\gamma}^2 \xi (3 - 4\xi)}{\pi \rho R^3} \left( \frac{3}{c_i^2} - \frac{2}{c_i^2} \right) \left[ (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 - \frac{1}{3} \right] \equiv \frac{A}{R^3} \left[ (\hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}})^2 - \frac{1}{3} \right]
\tag{10}
\]

Using the relation \( \eta_i^2 = 4\xi^2 \tilde{\gamma}^2 / 15 \), we can write the constant \( A \) as

\[
A \equiv \frac{15\eta_i^2}{4\pi \rho} \left( \frac{3}{\xi} - 4 \right) \left( \frac{3}{c_i^2} - \frac{2}{c_i^2} \right)
\tag{11}
\]

We can observe from Eq. (10) that the average of \( \langle U_{12} \rangle_{\hat{\mathbf{t}}_2} \) over \( \mathbf{\hat{n}} \) gives zero, as one would expect. Moreover, the TLSs placed along the direction \( \hat{\mathbf{t}}_1 \) have an average effect opposite to that of the TLSs placed in a plane perpendicular to \( \hat{\mathbf{t}}_1 \); the plane that goes through the TLS 1. In other words, if \( \langle U_{12} \rangle_{\hat{\mathbf{t}}_2} > 0 \) at \( \hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}} = \pm 1 \), then \( \langle U_{12} \rangle_{\hat{\mathbf{t}}_2} < 0 \) at \( \hat{\mathbf{t}}_1 \cdot \mathbf{\hat{n}} = 0 \), and viceversa.

For concrete calculations of the interaction energy, we have to evaluate the parameter \( \xi \). This is a material dependent parameter and is determined by the ratio \( \eta_i / \eta_i \) [5],

\[
\frac{4\gamma_i^2}{\eta_i^2} = \frac{15}{\xi^2} - \frac{40}{\xi} + 32.
\tag{12}
\]
Therefore, there is an ambiguity in determining $\xi$. For each material, i.e. for every physically possible value of $\gamma_l/\gamma_t$, there are two solutions of Eq. (12), $\xi_1$ and $\xi_2$, and up to now we have not been able to determine which one is physically relevant and which one is not; so in the calculations we have to consider both of them. But now from Eq. (12) we notice that

$$\frac{3}{\xi_{1,2}} - 4 = \pm \frac{2}{5} \sqrt{15 \frac{\gamma^2_l}{\gamma^2_t} - 20},$$

so for both values of $\xi$, the modulus of $A$ is the same and the ambiguity remains only in the sign:

$$A \equiv \pm \frac{3\gamma^2_l}{2\pi \rho} \sqrt{15 \frac{\gamma^2_l}{\gamma^2_t} - 20} \left( \frac{3}{c^2_l} - \frac{2}{c^2_t} \right).$$

One of the most studied glassy materials is the amorphous silica (a-SiO$_2$). For this material $\gamma_l = 1.04$ eV and $\gamma_t = 0.65$ eV [6], which give $\xi_1 \approx 1.31$ and $\xi_2 \approx 0.52$. Using also the other physical parameters, $c_l = 5.8$ km/s, $c_t = 3.8$ km/s, and $\rho = 2200$ kg/m$^3$ [6], we obtain $|A| \approx 9.35$ eVÅ$^3$.

As another example we take Epoxy. For this material, $\gamma_l = 0.35$ eV, $\gamma_t = 0.65$ eV, $c_l = 3.25$ km/s, $\rho = 1200$ kg/m$^3$, while $c_t$ we approximate by the quasi-universal relation for glasses, $c_t \approx c_l/1.65$ [6]. Plugging all these parameters into the expression for $A$, we get $|A| \approx 7.65$ eVÅ$^3$.

3 Conclusions

We used the model of Ref. [5] to calculate the TLS-TLS interaction in amorphous glassy materials. In this model there are spatial directions associated to all the TLS. Therefore, the TLS-TLS interaction Hamiltonian that we obtained here depends on the orientations of the two TLSs that interact and also on the direction of the line that connects them. The angular dependence of the interaction allows for a more detailed treatment of the TLS-TLS interaction in glassy materials and a deeper understanding of the low temperature properties of these materials.

Due to the isotropy of amorphous solids, one can assume that also the TLS orientations are isotropically distributed. We used this assumption to average over the orientations of one of the TLSs (say TLS 2) and we obtained a much simpler (average) interaction Hamiltonian, but which was still dependent on the angle between the orientation of the remaining TLS (the TLS1) and the line connecting TLS1 to TLS2. Based on the knowledge of $\gamma_l$ and $\gamma_t$, one can determine the interaction strength, up to its sign. Moreover, it was interesting to note that the interaction changes sign when going from the lateral direction ($\hat{t}_1 \cdot \hat{n} = 0$) to forward or backward direction ($\hat{t}_1 \cdot \hat{n} = \pm 1$).
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