3. Two Level Systems in Glasses. Model, Interaction, Spectral Diffusion and Interaction Stimulated Dynamics.

The main target of the consideration below is to study the delocalization in systems with the long-range excitation transfer. This concept will be illustrated on the example of two level systems (TLS) in amorphous solids. We start with the brief review of TLS model and related properties, and then will proceed to study various interaction effects.

## a. Model of Two Level Systems

The low temperature properties of amorphous solids below few Kelvin are rather universal. As was discovered by Zeller and Pohl in 1971 the behavior of specific heat and thermal conductivity in a number of dielectric glasses was quite similar to each other, namely specific heat shows linear temperature dependence, while the thermal conductivity is proportional to the squared temperature. Also the specific heat shows logarithmic dependence on the time of measurements.

This behavior is surprising for dielectric materials, where the low temperature properties are associated with the vibrational modes (phonons). Specific heat associated with phonons is proportional to the third power of the temperature which takes place in crystalline counterparts of glasses. Thermal conductivity is much bigger in crystalline materials. Therefore one can expect that there exist additional elementary excitations responsible for the observed universal behavior.

The hypothesis leading to the successful explanation of the majority of anomalous properties in amorphous solids was put forward in 1972 by Anderson, Halperin and Warma and independently Phillips. They suggested that there exist tunneling two level systems formed by atoms or groups of atoms undergoing tunneling motion between two



Fig. 3.1. Tunneling two level systems and definition of its parameters

close energy minima (Fig. 3.1). Each two level system (TLS) can be represented as two wells separated by the potential barrier. The position of particle can be described using a spin  $\frac{1}{2}$  operator, namely S<sup>z</sup>=1/2 for the left well and  $S^{z}=-1/2$  for the right well. The asymmetry of wells is characterized by the parameter  $\varDelta$  and the tunneling coupling of two separate states is characterized by the tunneling amplitude  $\Delta_0$ . The energy of TLS can be expressed using the Hamiltonian

$$h = -\Delta S^{z} - \Delta_{0} S^{x}. \qquad (3.1)$$

Remember that  $S^z$  and  $S^x$  are spin-1/2 –operators that can be expressed using Pauli matrices as

$$S^{z} = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad S^{x} = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}.$$
 (3.2)

Parameters  $\Delta$  and  $\Delta_0$  are different from TLS to TLS and one can characterize them by the distribution function  $P(\Delta, \Delta_0)$ . The right choice of this function is the critical issue for the model success. One can expect that the distribution of the parameter  $\Delta$  is nearly uniform because the thermodynamics of glasses at low temperature  $T \sim 1K$  is defined by energy range  $k_B T \sim 0.0001 eV$  which is very small compared to characteristic energies of order of Debye energy of larger to expect remarkable changes in the distribution function. The distribution of the tunneling parameter  $\Delta_0$  is more complicated. One can assume that since the tunneling amplitude is exponentially sensitive to the parameters of the potential barrier it will result in the logarithmically uniform distribution of  $\Delta_0$ . Indeed, expressing  $\Delta_0 = \exp(-a)$  where a is some parameter having the distribution g(a) and characterizing the potential barrier separating two minima Fig. 3.1. Then one can represent the distribution of tunneling amplitudes in the form  $P(\Delta_0) = \int_{-\infty}^{+\infty} g(a) da \delta(\Delta_0 - \exp(-a)) = \frac{g(\ln(\Delta_0))}{\Delta_0}.$  This leads to the following anzats for

any reasonable distribution function g(a) which depends on the tunneling amplitude much weaker then the factor  $1/\Delta_0$ 

$$P(\Delta, \Delta_0) = \frac{P_0}{\Delta_0}.$$
(3.3)

This distribution can be used to explain the anomalous behavior of the specific heat and the thermal conductivity at low temperature described above. Note that this distribution represents the TLS density in asymmetry energy, tunneling amplitude and space so that the probability to find TLS in the given volume element dV, with asymmetry between ( $\Delta$ ,  $\Delta$ +d $\Delta$ ), and tunneling amplitude in the domain ( $\Delta_0$ ,  $\Delta_0$ +d $\Delta_0$ ) is given by

$$\frac{P_0 d\Delta d\Delta_0}{\Delta_0} dV. \tag{3.3A}$$

Consider first the specific heat. For an individual two level system it can be expressed using the eigenenergies of the Hamiltonian Eq. (3.1), which can be obtained for instance introducing the transformation in the pseudospin space as

$$\widetilde{S}^{z} = \frac{\Delta}{E}S^{z} + \frac{\Delta_{0}}{E}S^{x}, \quad \widetilde{S}^{x} = -\frac{\Delta_{0}}{E}S^{z} + \frac{\Delta}{E}S^{x}, \quad \widetilde{S}^{y} = S^{y},$$

$$E = \sqrt{\Delta^{2} + \Delta_{0}^{2}}.$$
(3.4)

Then the Hamiltonian Eq. (3.1) can be rewritten as

$$\hat{h} = -E\tilde{S}^{z}.$$
(3.5)

Since eigenvalues of the spin  $\frac{1}{2}$  operator are always  $\pm 1/2$  one can express energies of TLS as  $\pm E/2$ . Accordingly average energy can be defined using the Botlzman distribution

$$\langle E \rangle = \frac{\frac{E}{2} \exp\left(\frac{E}{2T}\right) - \frac{E}{2} \exp\left(-\frac{E}{2T}\right)}{\left(\exp\left(\frac{E}{2T}\right) + \exp\left(-\frac{E}{2T}\right)\right)}.$$
(3.6)

The specific heat can be defined as the derivative of the average energy over the temperature. This yields

$$c_E = \frac{\left(\frac{E}{2T}\right)^2}{\cosh^2\left(\frac{E}{2T}\right)}.$$
(3.7)

This expression should be summed over all two level systems and divided by the system volume, which is equivalent to averaging with the distribution Eq. (3.3)

$$C = P_0 \int_{-\infty}^{+\infty} d\Delta \int_{0}^{+\infty} \frac{d\Delta_0}{\Delta_0} \frac{\left(\frac{E}{2T}\right)^2}{\cosh^2\left(\frac{E}{2T}\right)}.$$
(3.8)

It is more convenient to change integration variables to *E* and  $\Delta_0$  so Eq. (3.8) can be rewritten as

$$C = P_0 \int_0^{+\infty} \frac{d\Delta_0}{\Delta_0} \int_{\Delta_0}^{+\infty} \frac{EdE}{\sqrt{E^2 - \Delta_0^2}} \frac{\left(\frac{E}{2T}\right)^2}{\cosh^2\left(\frac{E}{2T}\right)} =$$

$$= P_0 \int_0^{+\infty} \frac{EdE\left(\frac{E}{2T}\right)^2}{\cosh^2\left(\frac{E}{2T}\right)} \int_{\Delta_{0\,\min}}^E \frac{d\Delta_0}{\Delta_0\sqrt{E^2 - \Delta_0^2}} =$$

$$= P_0 \int_0^{+\infty} \frac{dE\left(\frac{E}{2T}\right)^2}{\cosh^2\left(\frac{E}{2T}\right)} \ln\left(\frac{2E}{\Delta_{0\,\min}}\right)$$

$$= 2P_0 T \ln\left(\frac{4T}{\Delta_{0\min}}\right) \int_0^{+\infty} \frac{dxx^2}{\cosh^2(x)} + 2P_0 T \int_0^{+\infty} \frac{dxx^2}{\cosh^2(x)} \ln(x).$$
(3.9)

The low limit for integration over  $\Delta_0$  is taken to be finite because otherwise the number of TLS diverges. Evaluating integrals in Eq. (3.9) we got

$$C = 1.645P_0 T \ln\left(\frac{4T}{\Delta_{0\min}}\right) + 0.58P_0 T.$$
(3.10)

This result explains the linear temperature dependence of the glass specific heat. The logarithmic time dependence can be understood if we notice that at experimental time t only TLS having relaxation time smaller than t should be count in any equilibrium properties. Other TLS are not in equilibrium and cannot contribute to the specific heat. Since the relaxation time of TLS increases with decreasing  $\Delta_0$  as  $C/\Delta_0^2$  one can define the minimum tunneling amplitude as  $\Delta_{0min}=(c/t)^{1/2}$  and substituting this expression into Eq. (3.10) we obtain the logarithmic time dependence as in the experiments

$$C = 0.822P_0 T \ln\left(\frac{16T^2 t}{c}\right) + 0.58P_0 T.$$
(3.11)

To describe the thermal conductivity we need to introduce the interaction of TLS with the bath that is represented by phonons in dielectric glasses. We assume that phonons are responsible for the thermal conductivity and their absorption by TLS is the main mechanism of their scattering. Then if the phonon decay rate for thermal phonons (with energy  $\hbar\omega = k_B T$ ) is given by  $\gamma$  one can estimate the thermal conductivity as

$$\kappa = C \frac{v^2}{3\gamma},\tag{3.12}$$

where

$$C = 2.63 \cdot \frac{(k_B T)^3}{(\hbar v)^3}$$
(3.13)

is the phonon specific heat of the corresponding spectral branch and v is the speed of sound.

To find  $\gamma$  consider the rate of sound wave absorption by TLS, where the sound wave is expressed as

$$u_a(\mathbf{r},t) = e_a u_0 \cos(\mathbf{q}\mathbf{r} - \omega t),$$
  

$$\omega = qv,$$
(3.14)

where q and  $\omega$  are wavevector and frequency of the sound wave under consideration,  $u_0$  is its amplitude and e is the polarization vector which is parallel to q for longitudinal mode and perpendicular to q for two transverse modes. The rate of absorption is defined as the rate of energy absorption divided by the sound wave energy, which can be expressed as

$$E = \hbar \omega n, \quad n = u_0^2 \frac{MN\omega}{2\hbar} \to E = \frac{M\omega^2 u_0^2}{2}.$$
(3.15)

where M is the mass of the elementary cell and N is the number of cells in the crystal.

TLS interacts not with the wave Eq. (3.14) itself but with its gradients because of the translational symmetry of the system. If the wave is coordinate independent, then there is no potential energy change associated with it. Therefore the interaction of wave and TLS can be written in the form

$$\hat{V}_{ext} = -S_i^z \sum_{a,b=x,y,z} \gamma_{ab} \frac{\partial u_a}{\partial x_b} = -S_i^z \sum_{a,b=x,y,z} \gamma_{ab} q_b e_a u_0 \sin(\mathbf{qr} - \omega t), \qquad (3.16)$$

where  $\gamma_{ab}$  is the strain tensor characterizing the deformation associated with TLS transitions. The effect of vibration on the tunneling amplitude is neglected because the essential change in TLS asymmetry occurs when the interaction strength reaches the thermal energy T, while the essential change in TLS tunneling amplitude takes place when the energy change is of order of  $\hbar\omega_0$  which is the quantization energy of harmonic oscillator in the TLS potential well (Fig. 3.1). Since this energy is of order of the Debye energy it remarkably exceeds the thermal energy for T < 1K and therefore we can ignore this effect because all relevant interactions are smaller than the thermal energy.

One can use the Fermi golden rule to derive the absorption coefficient for interaction Eq. (3.16). It can be expressed as the difference of energy absorption and

energy emission rates between its ground TLS state  $\tilde{S}^z = 1/2$  and its excited state  $\tilde{S}^z = -1/2$ . The rate of TLS transition accompanied by absorption of the energy  $\hbar \omega$  is given by

$$r_{+} = P_{-} \frac{2\pi}{4\hbar} \Big| \langle \tilde{S}^{z} = 1/2 | S^{z} | \tilde{S}^{z} = -1/2 \rangle \Big|^{2} \Big| \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} u_{0} \Big|^{2} \delta(E - \hbar\omega), \quad (3.17)$$

while the rate of transitions accompanied by energy emission reads

$$r_{-} = P_{+} \frac{2\pi}{4\hbar} \Big| < \tilde{S}^{z} = 1/2 |S^{z}| \tilde{S}^{z} = -1/2 > \Big|^{2} \Big| \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} u_{0} \Big|^{2} \delta(E - \hbar\omega), \quad (3.18)$$

and  $P_{-}$  and  $P_{+}$  are probabilities to find the given TLS in its ground or excited states, respectively

$$P_{-} = \frac{e^{\frac{E}{kT}}}{e^{\frac{E}{kT}} + 1}, \quad P_{-} = \frac{1}{e^{\frac{E}{kT}} + 1}.$$
(3.19)

Substituting Eq. (3.19) into Eqs. (3.17), (3.18), using definition Eq. (3.4) and subtracting emission rate from absorption rate we got the following expression for the energy absorption rate

$$r_{s} = \pi \frac{\gamma_{\mu}^{2} \omega^{3}}{8 v_{\mu}^{2}} \frac{\Delta_{0}^{2}}{E^{2}} u_{0}^{2} \tanh\left(\frac{E}{2kT}\right) \delta(E - \hbar \omega),$$

$$\gamma_{\mu}^{2} = \frac{1}{q^{2}} \left| \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} \right|^{2}.$$
(3.20)

Index  $\mu$  enumerates vibrational modes and it can be either *l* for longitudinal mode or *t* for two transverse modes.

This result should be summed over all TLS in the sample which is equivalent to the following averaging

$$r_{tot} = \frac{\pi \langle \gamma_{\mu}^2 \rangle \omega^3 P_0 V}{8 v_{\mu}^2} \int_0^{+\infty} dE \int_0^E \frac{E d\Delta_0}{\sqrt{E^2 - \Delta_0^2} \Delta_0} \frac{\Delta_0^2}{E^2} u_0^2 \tanh\left(\frac{E}{2kT}\right) \delta(E - \hbar\omega).$$
(3.21)

The integrals can be evaluated analytically and we get

$$r_{tot} = \frac{\pi \langle \gamma_{\mu}^2 \rangle \omega^3 P_0 V u_0^2}{8 v_{\mu}^2} \tanh\left(\frac{\hbar \omega}{2kT}\right).$$
(3.22)

Using Eq. (3.15) one can express the phonon decay rate as

$$\gamma = \frac{1}{\tau} = \frac{\pi}{4} P_0 \frac{\langle \gamma_\mu^2 \rangle}{\rho v_\mu^2} \omega \tanh\left(\frac{\hbar\omega}{2kT}\right).$$
(3.23)

The main contribution to thermal conductivity is associated with thermal phonons characterized by the thermal energy  $\hbar\omega$ ~kT. The decay rate of thermal phonons scales as

$$\gamma_T \approx \frac{\pi}{4} P_0 \frac{\left\langle \gamma_\mu^2 \right\rangle}{\rho v_\mu^2} \frac{kT}{\hbar}.$$
(3.24)

Substituting this result into Eq. (3.12) we obtain the behavior of the thermal conductivity

$$k = 2.63 \cdot \frac{\left(k_B T\right)^2}{\left(\hbar v\right)^3} v^2 \frac{4\hbar}{\pi P_0 \frac{\left\langle \gamma_\mu^2 \right\rangle}{\rho v_\mu^2}}.$$
(3.25)

Thus the model of two-level systems successfully explains the low-temperature behavior of the specific heat and the thermal conductivity in amorphous solids. It is interesting that the dimensionless ratio of phonon absorption rate Eq. (3.23) and phonon frequency  $\omega$  or the ratio of the phonon mean free path to the phonon wavelength is expressed by the dimensionless parameter

$$\frac{\pi}{4}P_0\frac{\left\langle\gamma_{\mu}^2\right\rangle}{\rho v_{\mu}^2}.$$
(3.26)

This parameter is universal in various glasses and its universality was used by theorists to suggest some models for TLS formation.

Other parameters often used to characterize the universal low temperature glassy behavior are the sound attenuation (or internal friction) at  $\hbar\omega < < kT$  and temperature dependent velocity of sound. Similar behavior takes place for dielectric constant and dielectric losses.

The absorption described by Eq. (3.23) is called resonant absorption because it is associated with the transition of TLS having the energy equal to the sound energy quantum. If  $\hbar\omega < <kT$  this mechanism is substantially suppressed by the population difference factor  $\tanh\left(\frac{\hbar\omega}{2kT}\right)$  (of order or less than  $10^{-4}$ ). At sufficiently small frequency the absorption nevertheless approaches Eq. (3.24) due to the different, so called

the absorption nevertheless approaches Eq. (3.24) due to the different, so called relaxational, mechanism. Qualitatively this mechanism can be described as following. The external field Eq. (3.16) changes the equilibrium population difference of TLS. Since the relaxation of some TLS is slow their population difference deviates strongly from equilibrium and therefore they absorb sound. To characterize this absorption one has to compute TLS relaxation time caused by their interaction with phonons.

The relaxation time can be computed assuming that TLS is in its ground or excited state and computing the time of absorption or emission of phonon, accompanied by TLS transition between two states. The interaction of the given TLS with phonons can be taken in the form of Eq. (3.16)

$$\hat{V} = -S_i^z \sum_{a,b=x,y,z} \gamma_{ab} \frac{\partial u_a}{\partial x_b} = S_i^z \sum_{a,b=x,y,z} \gamma_{ab}^\mu q_b e_a^\mu \sqrt{\frac{\hbar}{2M\omega_\mu N}} \left( b_{q\mu}^+ - b_{q\mu} \right).$$
(3.27)

TLS transition rates between its ground and excited states and backwards can be evaluated similarly to Eqs. (3.17), (3.18)

$$r_{+} = \frac{\pi}{4\hbar} \frac{\Delta_{0}^{2}}{E^{2}} \frac{V\hbar}{2MN} \sum_{\mu} \int \frac{d\mathbf{q}}{(2\pi)^{3} \omega_{\mu}} \left| \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} \right|^{2} \delta(E - \hbar \omega_{q\mu}) \frac{1}{\exp\left(\frac{\hbar \omega_{q\mu}}{kT}\right) - 1}, \quad (3.28)$$

$$r_{-} = \frac{\pi}{4\hbar} \frac{\Delta_{0}^{2}}{E^{2}} \frac{V\hbar}{2MN} \sum_{\mu} \int \frac{d\mathbf{q}}{(2\pi)^{3} \omega_{\mu}} \left| \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} \right|^{2} \delta(E - \hbar \omega_{q\mu}) \frac{\exp\left(\frac{\hbar \omega_{q\mu}}{kT}\right)}{\exp\left(\frac{\hbar \omega_{q\mu}}{kT}\right) - 1}.$$
(3.29)

Evaluation of integrals for both rates yields

$$r_{+} = \frac{1}{16\pi\hbar^{4}} \frac{\Delta_{0}^{2}E}{\exp\left(\frac{E}{kT}\right) - 1} \sum_{\mu} \frac{\gamma_{\mu}^{2}}{\rho v_{\mu}^{5}},$$
(3.30)
$$r_{-} = \frac{1}{16\pi\hbar^{4}} \frac{\Delta_{0}^{2}E \exp\left(\frac{E}{kT}\right)}{\exp\left(\frac{E}{kT}\right) - 1} \sum_{\mu} \frac{\gamma_{\mu}^{2}}{\rho v_{\mu}^{5}}.$$
(3.31)

Finally the relaxation rate of TLS can be defined using the Bloch equations for probabilities to occupy ground and excited states

$$\frac{dP_g}{dt} = -r_+ P_g + r_- P_{ex} = -\frac{dP_{ex}}{dt}.$$
(3.32)

Using the identity  $P_g+P_{ex}=1$  we express the relaxation in term of the single rate  $r=r_++r_$  $dP_a$ 

$$\frac{dr_{g}}{dt} = -rP_{g} + r_{-},$$

$$\frac{dP_{ex}}{dt} = -rP_{ex} + r_{+}.$$

$$r = \frac{\Delta_{0}^{2}E}{16\pi\hbar^{4}} \operatorname{coth}\left(\frac{E}{2kT}\right) \sum_{\mu} \frac{\gamma_{\mu}^{2}}{\rho v_{\mu}^{5}}.$$
(3.33)

At this point we are ready to derive the expression for the relaxational absorption. The equilibrium population of two level systems changes in the presence of the external field Eq. (3.16) as

$$\Delta P_{eq} = P_g(t) - P_{ex}(t) = \tanh\left(\frac{\sqrt{\Delta_0^2 + \left[\Delta - \sum_{a,b=x,y,z} \gamma_{ab} q_b e_a u_0 \sin(\omega t)\right]^2}}{2kT}\right).$$
(3.34)

The rate of the energy absorption from the external field can be expressed as the average time derivative of the interaction Hamiltonian Eq. (3.16)

$$\left\langle \frac{\partial \hat{V}_{ext}}{\partial t} \right\rangle_{s} = \omega \left\langle S^{z} \right\rangle \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} u_{0} \cos(\omega t) = \frac{\Delta \omega}{E} \Delta P(t) \sum_{a,b=x,y,z} \gamma_{ab} q_{b} e_{a} u_{0} \cos(\omega t), \qquad (3.35)$$

where the average population difference  $\Delta P$  is defined by the Bloch's equation  $d\Delta P$ 

$$\frac{d\Delta H}{dt} = -r(\Delta P - \Delta P_{eq}). \tag{3.36}$$

Within the linear response theory we need to hold only linear term in  $u_0$  in  $\Delta P_{eq}$ 

$$\Delta P_{eq} \approx \tanh\left(\frac{E}{2kT}\right) - \frac{\Delta}{2kTE\cosh^2\left(\frac{E}{2kT}\right)} \sum_{a,b=x,y,z} \gamma_{ab} q_b e_a u_0 \sin(\omega t).$$
(3.37)

Then the solution to Eq. (3.36) linear in  $u_0$  can be expressed as

$$\Delta P(t) = \tanh\left(\frac{E}{2kT}\right)$$

$$-\frac{r^{2}}{r^{2}+\omega^{2}}\frac{\Delta}{2kTE\cosh^{2}\left(\frac{E}{2kT}\right)}\sum_{a,b=x,y,z}\gamma_{ab}q_{b}e_{a}u_{0}\sin(\omega t)$$

$$+\frac{r\omega}{r^{2}+\omega^{2}}\frac{\Delta}{2kTE\cosh^{2}\left(\frac{E}{2kT}\right)}\sum_{a,b=x,y,z}\gamma_{ab}q_{b}e_{a}u_{0}\cos(\omega t).$$
(3.38)

This expression should be substituted into Eq. (3.35) and then the contribution of the given TLS to the relaxational absorption can be expressed as

$$\left\langle \frac{\partial \hat{V}_{ext}}{\partial t} \right\rangle_{s} = \frac{1}{4} u_{0}^{2} \left( \frac{\Delta}{E} \right)^{2} \frac{r \omega^{4}}{r^{2} + \omega^{2}} \frac{1}{2 v_{\mu}^{2} k T \cosh^{2} \left( \frac{E}{2kT} \right)} \left( \sum_{a,b=x,y,z} \gamma_{ab} e_{\mu b} e_{a} \right)^{2}.$$
(3.39)

This contribution should be divided by the mode vibrational energy Eq. (3.15) and summed over all TLS. This yields the sound decay rate

$$\gamma = \omega \frac{\gamma_{\mu}^{2}}{4\rho v_{\mu}^{2}} \frac{P_{0}}{kT} \int_{0}^{+\infty} dE \frac{E}{\cosh^{2}\left(\frac{E}{2kT}\right)} \int_{0}^{E} \frac{d\Delta_{0}}{\Delta_{0}\sqrt{E^{2} - \Delta_{0}^{2}}} \frac{\omega r}{1 + \omega^{2}r^{2}} \frac{E^{2} - \Delta_{0}^{2}}{E^{2}}.$$
(3.40)

It is convenient to replace  $\Delta_0$  with the new variable  $x=\Delta_0/E$ . Then the integral Eq. (3.40) can be written as

$$\gamma = \omega \frac{\gamma_{\mu}^{2}}{4\rho v_{\mu}^{2}} \frac{P_{0}}{kT} \int_{0}^{+\infty} dE \frac{1}{\cosh^{2}\left(\frac{E}{2kT}\right)} \int_{0}^{1} \frac{dx\sqrt{1-x^{2}}}{x} \frac{\omega Ax^{2}E^{3} \coth\left(\frac{E}{2T}\right)}{\omega^{2} + \left[Ax^{2}E^{3} \coth\left(\frac{E}{2T}\right)\right]^{2}}, \quad (3.41)$$
$$A = \frac{1}{16\pi\hbar^{4}} \sum_{\mu} \frac{\gamma_{\mu}^{2}}{\rho v_{\mu}^{5}}.$$

The expression under first integral is defined by the energies E of order of thermal energy. Then depending on the relationship between the frequency  $\omega$  and the maximum efficient transition rate  $r_{max} \sim AT^3$  we have two different behaviors. At low frequency

$$\omega \ll AT^3, \tag{3.42}$$

The integral in Eq. (3.41) can be evaluated because it is defined by the intermediate x << 1. After straightforward calculations we get

$$\gamma = \frac{1}{\tau} = \frac{\pi}{8} P_0 \frac{\left\langle \gamma_{\mu}^2 \right\rangle}{\rho v_{\mu}^2} \omega.$$
(3.43)

This expression is formally two times smaller than the one for the resonant absorption Eq. (3.23) in the limit of a very high frequency  $\hbar\omega$ >>kT.

In the opposite limit of high frequency or low temperature

$$\omega \gg AT^3, \tag{3.44}$$

One can also evaluate the integral Eq. (3.41) neglecting the second term in the denominator of Eq. (3.41). Then we get

$$\gamma = \omega P_0 \frac{\gamma_{\mu}^2}{3\rho v_{\mu}^2} \frac{AT^3}{\omega} \int_0^{+\infty} du \frac{u^3}{\sinh(u)} \approx 2\omega P_0 \frac{\gamma_{\mu}^2}{\rho v_{\mu}^2} \frac{AT^3}{\omega}.$$
(3.45)

The attenuation of sound is defined by the ratio  $\gamma/v_{\mu}$ . It is convenient to use the dimensionless parameter of internal friction defined as the ratio  $Q^{-1} = \gamma/\omega$ . This parameter is universal constant

$$Q_{\mu}^{-1} = \frac{\pi}{8} P_0 \frac{\left\langle \gamma_{\mu}^2 \right\rangle}{\rho v_{\mu}^2} \tag{3.46}$$

at larger temperatures

$$T > T_{\omega} \sim \left(\frac{A}{\omega}\right)^{1/3},\tag{3.47}$$

(for typical experimental frequency  $\omega \sim 10^4 s^{-1}$  the crossover temperature  $T_{\omega}$  is around 50mK) Eq. (3.43) and decreases at lower temperatures as  $T^3$ 

$$Q_{\mu}^{-1} \approx 2P_0 \frac{\gamma_{\mu}^2}{\rho v_{\mu}^2} \frac{AT^3}{\omega}.$$
 (3.48)

This behavior permits the direct measurements of the typical relaxation rate of thermal TLS having both energy and tunneling amplitude of order of the thermal energy.

The frequency dependence of the sound attenuation (internal friction) can be used to study the temperature and frequency dependence of the sound velocity, which is coupled to the internal friction by the Kramers-Kronig relationships

$$\frac{\Delta v_{\mu}^{2}}{v_{\mu}^{2}} = -\frac{2}{\pi} P.V. \int_{0}^{+\infty} \frac{Q_{\mu}^{-1}(\omega')\omega' d\omega'}{\omega'^{2} - \omega^{2}}.$$
(3.49)

The main contribution to Eq. (3.49) is defined by the domains where  $Q^{-1}$  is constant and  $\omega$ '> $\omega$  because of the logarithmic character of the integrals there. Then at low temperature the logarithmic temperature dependence comes from the domain  $kT/\hbar < \omega < \omega_{max}$  where  $\omega_{max}$  is some natural cutoff or TLS energies

$$\frac{\Delta v_{\mu}^{2}}{v_{\mu}^{2}} = -C \ln\left(\frac{\hbar\omega_{mx}}{kT}\right),$$

$$C = P_{0} \frac{\gamma_{\mu}^{2}}{\rho v_{\mu}^{2}}.$$
(3.50)

At higher temperatures  $T>T_{\omega}$  the domain  $\omega < \omega' < AT^3$  also contributes to the logarithmic temperature dependence of the velocity of sound so one can express it as

$$\frac{\Delta v_{\mu}^{2}}{v_{\mu}^{2}} = -C \ln\left(\frac{\hbar\omega_{mx}}{kT}\right) - \frac{1}{2}C \ln\left(\frac{AT^{3}}{\omega}\right).$$
(3.51)

Thus theory predicts that the sound velocity has a maximum in its temperature dependence at  $T \sim T_{\omega}$ . At lower temperatures it decreases logarithmically with decreasing the temperature. The slope of the logarithmic temperature dependence is C at low temperatures, while above  $T_{\omega}$  the sound velocity decreases with increasing the temperature logarithmically with the slope -C/2.

Both the broad domain of temperature independent internal friction and the logarithmic temperature dependence of the velocity of sound have been found experimentally. This behavior is considered as the evidence of the existence of TLS in particular material.

In metal glasses the TLS relaxation rate behaves differently. It shows linear temperature dependence because the phase volume of electron-hole pairs interacting with TLS scales linearly with the temperature in contrast with the phonon phase volume  $T^3$ . The TLS relaxation rate can be expressed as

$$r = B \frac{\Delta_0^2}{E^2} \frac{kT}{\hbar},\tag{3.52}$$

where the constant B usually belongs to the domain between 0.1 and 1. This behavior of relaxation rate changes qualitatively the sound velocity temperature dependence. At low temperature  $T < \hbar \omega / k$  the temperature dependence of sound velocity still obeys the law Eq. (3.50) while at higher temperatures sound velocity still increases with the temperature although the slope of the logarithmic dependence changes to C/2. The internal friction at higher temperatures follows Eq. (3.48), while at low temperatures it depends linearly on temperature as

$$Q_{\mu}^{-1} \approx P_0 \frac{\gamma_{\mu}^2}{\rho v_{\mu}^2} \frac{BT}{\omega}.$$
(3.53)

This universal behavior was observed in various metal glasses.

The dielectric losses  $\varepsilon$ ' and dielectric constant  $\varepsilon$ ' behave quite similarly because TLS possess dipole moments. The derivation can be made in the similar way to the internal friction and speed of sound. Below I give only answers, which are qualitatively transparent. For higher temperatures T>T<sub> $\omega$ </sub> Eq. (3.47) dielectric losses are temperature independent and can be expressed as

$$\frac{\varepsilon''}{\varepsilon} = \frac{\pi}{8} C_{\varepsilon}, \quad C_{\varepsilon} = P_0 \frac{\mu^2}{\varepsilon}.$$
(3.54)

Here  $\mu^2$  is the characteristic average squared dipole moment of TLS. Dielectric constant shows logarithmic temperature dependence defined by slightly modified Eq. (3.51)

$$\frac{\Delta\varepsilon}{\varepsilon} = C_{\varepsilon} \ln\left(\frac{\hbar\omega_{mx}}{kT}\right) + \frac{1}{2}C_{\varepsilon} \ln\left(\frac{AT^{3}}{\omega}\right).$$
(3.55)

The sign of temperature dependence is opposite to that for the sound velocity. This is not surprising because the temperature dependence of the speed of light under these conditions is identical to that for the speed of sound because the speed of light is inversely proportional to  $\epsilon^{1/2}$ . At very low temperatures T<T<sub> $\omega$ </sub> we get

$$\frac{\Delta\varepsilon}{\varepsilon} = C_{\varepsilon} \ln\left(\frac{\hbar\omega_{mx}}{kT}\right),$$

$$\frac{\varepsilon''}{\varepsilon} \approx 2C_{\varepsilon} \frac{AT^{3}}{\omega}.$$
(3.56)

Thus the temperature dependence of the dielectric constant is similar to the one for the negative sound velocity. Dielectric constant has a minimum at  $T \sim T_{\omega}$ . The slope of logarithmic temperature dependence at high temperature side should be approximately  $\frac{1}{2}$  of the slope on the low temperature side.

Further investigation of two level systems reveals other interesting effects. Echo and hole burning experiments in glasses clearly demonstrated that the TLS decoherence rate associated with the phase of its quantum state cannot be described using the relaxation rate Eq. (3.33). The real decoherence rate is much larger and has a different temperature dependence. Analysis of TLS decoherence cannot be performed without the consideration of TLS long-range  $1/R^3$  interaction. Such interaction exists because TLS possess dipole moments and the interaction with phonons also results in similar  $1/R^3$  interaction. This consideration will be made in the next lecture.

Literature

- 1. Zeller and Pohl in 1971
- 2. Anderson, Halperin, Warma, 1972
- 3. Phillips, 1972
- 4. Arnold and Hunklinger 1975 (?)

Problem set

- 1. Using the definition of spin operators Eq. (3.2) find eigenvalues and eigenstates of the Hamiltonian Eq. (3.1) directly diagonalizing it.
- 2. According to the semiclassical theory the tunneling amplitude can be expressed as the following function of barrier height U and width a  $\Delta_0 = \Delta_* \exp\left(-\frac{2a\sqrt{mU}}{\hbar}\right)$

where  $\Delta_*$  is the preexponential factor and m is the mass of tunneling particle which we both set constant. Then assume that either a or U are uniformly distributed is some broad domain calculate the distribution function and compare it with Eq. (3.3). Do the same assuming both *a* and *U* uniformly distributed.

- 3. Verify that the redefinition of spin operators (3.4) conserves their commutation rules.
- 4. Derive kinetic formula Eq. (3.12). Hint: First derive the estimate for the phonon diffusion coefficient D~lv/3 and then express the thermal conductivity through energy diffusion.