# Weakly coupled tunneling systems in mixed crystals

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(Received 5 November 2001; revised manuscript received 1 July 2002; published 30 October 2002)

The low-temperature behavior of imperfect crystals like KCl with a few ppm of substitutional defects like $OH^-$ or $Li^+$ is dominated by the dynamics of these defects. At low concentrations the defects are usually described as isolated systems. We find that in typical experimental situations even at concentrations of only one ppm the defects are not independent of each other. Instead they form pairs. For weak coupling, bulk quantities like the dielectric constant are hardly affected, but the total moments (electric and elastic) of the pairs of tunneling systems depend on the relative angles between the tunneling systems. Since these angles take several discrete values, there is a set of Rabi frequencies, relaxation rates, and phase coherence times, affecting echo experiments.

DOI: 10.1103/PhysRevB.66.134107 PACS number(s): 61.72.Ji, 66.35.+a, 03.65.Yz

## I. INTRODUCTION

The low-temperature properties of alkali halides may be significantly modified by the presence of substitutional impurities, such as $Li^+$, $CN^-$, or $OH^-$. A few ppm of these defects are sufficient to change the thermal behavior and the elastic and electric response below a few Kelvin.

Regarding the polar molecules $CN^-$ and $OH^-$, the point symmetry of the impurity site gives rise to several equivalent orientations; corresponding off-center positions arise for the small lithium ion. Quantum tunneling between these states results in a ground-state splitting of about 1 K. Since the number density of such tunneling states exceeds, even at low concentrations, that of small-frequency phonon modes of the host crystal, the impurities govern the low-temperature properties of the material. The cubic symmetry of the fcc crystals favors six, eight, or twelve defect positions. The resulting energy spectra have been discussed in detail by Gomez *et al.*,[1] and agree well with the Schottky peak observed by Pohl and co-workers for various impurity systems at low doping.[2]

In defect crystals the importance of the coupling between the tunneling systems depends on the concentration of defects and ranges from weak (typically for a few ppm) to strong (typically for a few hundred or thousand ppm). Bulk quantities like the dielectric constant of such interacting tunneling systems are well described by a model of Würger.[3] But it is hard to gain information about the damping rates from that treatment.

The low-temperature properties of glasses are similarly dominated by tunneling systems. Although little is known about the detailed structure of these tunneling systems, the phenomenological standard tunneling model[4] (STM) provides a good description of most experimental facts.[5] In recent years, experiments at temperatures below about 200 mK showed clear deviations from the predictions of the STM[6] which are mainly attributed to weak interactions between the tunneling systems. Burin and Kagan[7] proposed a new relaxation mechanism due to weak interactions and the existence of a dipolar gap in the distribution of energy splittings. Despite the success of these investigations in explaining at least qualitatively most experimental features, there is still a lack of a more rigorous treatment of the dynamics of interacting tunneling systems.

The defects are often approximated by simple two-level systems (TLS's) simplifying the calculation especially when dealing with interacting tunneling defects.[3,8] In the case of tunneling defects in crystals with eight possible defect positions the static and the dielectric properties are usually well approximated whereas the elastic response differs drastically.[9,10] In Sec. IV C we shortly discuss tunneling defects taking into account their full structure, but for the sake of simplicity we will use the two-state approximation throughout the paper.

Since a TLS describes a particle moving in a double well potential there is a direction connected to each TLS, which we choose to be distributed homogeneously, and we assume that the electric moment is aligned with that direction. Simplifying the elastic moment to an *elastic dipole* we choose its direction to be homogeneously distributed as well. We additionally assume that the direction of the elastic moment is independent from the direction of the electric moment. This assumption is typically not justified but within the two-state approximation it is necessary to reveal effects discussed in the present paper which would be hidden if we assumed strictly aligned electric and elastic moments. In Sec. IV C we will justify our approach by showing that the mentioned effects are present for the defects taking their full structure into account.

The Hamiltonian of a TLS is given by

$$H_{\mathrm{TLS}} = -\frac{\Delta}{2}\sigma_x - \frac{F}{2}\sigma_z, \tag{1}$$

with the tunneling splitting $\Delta$ of the classical ground states and the Pauli matrices $\sigma_x$, $\sigma_y$, and $\sigma_z$. The matrix $\sigma_z$ corresponds to the real-space coordinate and $F$ is the energy bias between the ''left'' and ''right'' well given by external or internal fields. The internal fields can be divided into two subclasses depending on their source; defects which are static on the time scales of experiments yield static fields but, for example, fast (tunneling) defects or elastic waves in the medium generate dynamic fields and we have to take their dynamics into account. Neglecting static fields, we end up

 

with the Hamiltonian (9) in which we included interactions between tunneling defects. Each defect, therefore, sees a dynamic field generated by the other defects.

The coupling between the tunneling defects is modeled by a dipole-dipole type $J_{ij}\sigma_z^{(i)}\sigma_z^{(j)}$ term and the coupling constant $J_{ij}$ depends inversely on the cube of the distance between TLS's. Accordingly we expect a distribution of $P(J) \propto 1/J^2$ for homogeneously distributed defects. We thereby assume equal probabilities for both signs of the coupling to mimic the angular dependence of the dipole-dipole interaction. With rising concentration the mean value of the interaction increases.[3] At lowest concentration of a few ppm the interaction is usually negligible in measurements of the dielectric constant.

We want to investigate weakly interacting two-level systems coupled to a heat bath, a typical situation in a crystal with only a few ppm of tunneling defects. While such tunneling systems are usually described as isolated, we ask here, if a weak coupling leads to energy exchange between the tunneling systems and thus to a new relaxation mechanism. We show that there is no new relaxation mechanism. The coupled system separates into effective pairs. Each pair can be described as two effective uncoupled TLS and, then, even weak interactions result in a broad distribution of effective moments (electric, elastic) within the two-state approximation. Taking the full structure of the tunneling defects into account we obtain a discrete set of a few effective moments. Rates for energy relaxation or dephasing of a tunneling defect are given by the probability for an absorption or emission of a resonant phonon. If two TLS's forming a pair have identical energy splittings as is typically the case for crystals with only a few ppm of defects, a phonon with the same frequency can interact resonantly with both systems. Since the wavelength of such phonons is typically larger than the distance between the tunneling systems, the phonons cannot resolve the single moments but only the total moment. As the values of these total moments are broadly distributed, so are the relaxation rates (for energy and phase) of the systems instead of being peaked at the value which an isolated TLS would have. Such a distribution may show up best within echo experiments which are directly sensitive to the various rates.

Even if our model is not adequate to discuss interacting tunneling systems in glasses, since we neglected asymmetries (static internal fields), it can be viewed as a first step in that direction.

The paper is organized as follows. In the following section we give a short introduction to the Liouville formalism which gives the main theoretical framework for the subsequent discussion. In Sec. III we introduce the approximations used to calculate the dynamics of the defects coupled to a heat bath. In Sec. IV we investigate the dynamics of a coupled pair of TLS's and calculate the relaxation rates due to the interaction with the phonon bath. In Sec. V we show that within the assumption of weak coupling we can simplify the case of many interacting TLS's to a set of pairs. The last section contains a summary of our results and a discussion of their experimental relevance.

## II. LIOUVILLE FORMALISM

A convenient starting point is a formulation in Liouville space.[11] This is a linear space spanned by quantum-mechanical Hermitian operators in which each operator $\hat{A}$ is considered as a vector $|A)$. Superoperators $\mathcal{O}$ are introduced as mappings of the quantum-mechanical operators onto themselves. The von-Neumann equation for the statistical operator $\hat{\rho}(t)$ yields a simple example

$$\partial_t \hat{\rho}(t) = -i[\hat{H}(t), \hat{\rho}(t)] =: \mathcal{L}(t)|\rho(t)) \tag{2}$$

defining the Liouville superoperator $\mathcal{L}(t)$. We use $\hbar = 1$ throughout the paper. There are several possibilities to define a scalar product. In the following

$$(A|B) = \mathrm{Tr}\{\hat{A}^\dagger \hat{B}\} \tag{3}$$

is used. As in Hilbert space we can define a time evolution superoperator $\mathcal{U}(t,t')$ which obeys

$$\partial_t \mathcal{U}(t,t') = \mathcal{L}(t)\mathcal{U}(t,t') \tag{4}$$

with initial condition $\mathcal{U}(t,t) = \mathcal{I}$ and $\mathcal{I}|A) = |A)$. It is convenient to introduce at least two types of superoperators: the correlation operator $\mathcal{A}$ with

$$\mathcal{A}|X) := |\tfrac{1}{2}\{\hat{A}\hat{X} + \hat{X}\hat{A}\}), \tag{5}$$

and the response operator $\tilde{\mathcal{B}}$ with

$$\tilde{\mathcal{B}}|X) = |i[\hat{B}, \hat{X}]). \tag{6}$$

With this formal preparation we can express the thermodynamic average $\langle \hat{A} \rangle$, correlation functions $C_{A,B}(t,t')$ and, adding external fields to the Hamiltonian $\hat{H}(t) \to \hat{H} - h_B(t)\hat{B}$, the response functions $R_{A,B}(t,t')$ (within linear response):

$$\langle \hat{A} \rangle = (\mathbb{1}|\mathcal{A}|\rho),$$

$$C_{A,B}(t,t') = (1|\mathcal{A}\mathcal{U}(t,t')\mathcal{B}|\rho),$$

$$R_{A,B}(t,t') = \frac{\delta \langle \hat{A}(t) \rangle}{\delta h_B(t')} = (1|\mathcal{A}\mathcal{U}(t,t')\tilde{\mathcal{B}}|\rho). \tag{7}$$

The correlation and response functions are related by the fluctuation-dissipation theorem[11]

$$\chi''_{pp}(\omega) = 2\tanh(\beta\omega/2) C''_{pp}(\omega), \tag{8}$$

which connects the spectra $\chi''_{pp}(\omega)$ and $C''_{pp}(\omega)$. We define the Laplace transformation of $f(t)$ as $f(z) = i\int_0^\infty dt \exp(izt) f(t)$. Thus the spectra are given as the imaginary part of the Laplace transforms for $(z \to \omega)$ with $\omega$ real and $z$ in the upper complex half plane.

## III. MODEL

In this paper we address the problem of coupled two-level systems which are also coupled to a bath of phonons. Quite

generally, the heat bath may be reduced to a set of harmonic oscillators with linear coupling to the dipole operator $\sigma_z^{(i)}$ of the TLS's,[12] leading to

$$\hat{H}=H_S+H_{SB}+H_B,$$

$$H_S=-\sum_i \frac{\Delta_i}{2}\sigma_x^{(i)}-\sum_{i\neq j} J_{ij}\sigma_z^{(i)}\sigma_z^{(j)},$$

$$H_{SB}=\sum_i \sigma_z^{(i)}\cdot\sum_k \lambda_k^{(i)}(b_k+b_{-k}^{\dagger}),$$

$$H_B=\sum_k \omega_k b_k^{\dagger}b_k, \tag{9}$$

with the annilation (creation) operators for the phonons $b_k(b_k^{\dagger})$, respectively. The tunneling elements of tunneling defects in crystals are usually equal $\Delta_i=\Delta$. We consider the case of weak coupling between the tunneling defects assuming that the mean coupling $\overline{J(r)}$ between tunneling defects at the mean interparticle distance $\overline{r}$ is much smaller than the tunneling element $\Delta$ of the defects. The coupling between the phonons and the TLS's leads to damping for the latter. For crystals with sufficiently few defects, the time evolution of the phonons is not affected by the TLS's and the effect of the heat bath on the TLS's is entirely characterized by the generalized spectra

$$J^{(ij)}(\omega)=\frac{\pi}{2}\sum_k (\lambda_k^{(i)})^{\dagger}\lambda_k^{(j)}\delta(\omega-\omega_k), \tag{10}$$

where we assumed equal coupling constants for all defects. As long as the excitation energies of $H_S$ do not exceed a few Kelvin, only the low-energy part of the bath excitations is important. At these energies the spectral function $J^{(i=j)}(\omega)$ obeys a simple power-law behavior that is determined by the density of states $\Sigma_k\delta(\omega-\omega_k)$ and the frequency dependence of the coupling energies $\lambda_k^{(i)}$.

In insulating materials, low-frequency elastic waves provide the most efficient damping mechanism. The linear dispersion relation gives rise to a quadratic density of phonon states. Taking into account the frequency dependence $\lambda_k^{(i)}\propto\sqrt{\omega_k}$ due to the coupling of the elastic moment of a two-level system to the distortion of the host material due to the elastic waves, we obtain the well-known cubic law

$$J^{(i=j)}(\omega)=\pi\alpha_i\omega^3 \quad \text{with} \quad \alpha_i=\frac{3\gamma_i^2}{2\pi\hbar\rho v^5}, \tag{11}$$

where $\rho$ is the mass density of defects and $v$ is the sound velocity. $\gamma_i$ is the deformation potential of the defect $i$ and, therefore, the coupling constant between the TLS $i$ and the phonons.

There are various approaches for calculating the dissipative dynamics of single two-state systems coupled to a phonon bath.[12–15] The case of a pair of TLS's was considered by Terzidis and Würger[8] wherein they focused mainly on the dielectric constant. In the low-temperature limit, all of these approaches conclude that relaxation rates are governed by the direct or one-phonon process, resulting in weakly damped dynamics.

We address the question of how relaxation and dephasing rates are changed if we allow for small interactions between the TLS's. As we are only interested in the low-temperature results, we use a simple resummed perturbation series introduced by Horner and Würger[11,14] to deal with the coupling to phonons. Our main focus is how small interactions between the TLS's alter the damping rates compared to the rate of independent TLS's.

For example, in order to calculate the dipole-dipole correlation function $C_{pp}(t,t')$ we first integrate out all bath degrees of freedom. Formally we obtain

$$C_{pp}(t,t_0)=\int_{t_0}^{t}ds\int_{-\infty}^{t_0}ds'(1|\boldsymbol{\mathcal{P}}\boldsymbol{\mathcal{U}}_{\text{eff}}(t,s)\boldsymbol{\mathcal{P}}_{\text{ren}}(s,t_0,s')|\rho_{eq}) \tag{12}$$

with the statistical equilibrium operator of the pair $\rho_{eq}$ and the dipole superoperator $\boldsymbol{\mathcal{P}}$. We thereby assumed that at the time $t=-\infty$, both systems are decoupled and we can express the equilibrium as the direct product of the equilibrium operators of the pair and the bath. The following time evolution leads to a renormalization of the dipole operator $\boldsymbol{\mathcal{P}}_{\text{ren}}$. We give the above quantities in their lowest order.

Within the Laplace representation we get

$$U_{\text{eff}}(z)=-[\boldsymbol{\mathcal{U}}_S(z)^{-1}-M(z)]^{-1} \tag{13}$$

for the effective time evolution using the free time evolution $\boldsymbol{\mathcal{U}}_S(z)$ of the defects alone. To lowest order in $\boldsymbol{\mathcal{L}}_{SB}=-\tilde{H}_{SB}$ the memory kernel reads

$$M(t,t')=\text{Tr}\{\boldsymbol{\mathcal{L}}_{SB}U_0(t,t')\boldsymbol{\mathcal{L}}_{SB}\}_B, \tag{14}$$

where $U_0(t,t')$ equals the free time evolution. The trace $\text{Tr}\{\cdot\}_B$ only has to be taken with respect to the bath degrees of freedom.

The renormalized dipole operator is in lowest order

$$\boldsymbol{\mathcal{P}}_{\text{ren}}(s,t_0,s')=\boldsymbol{\mathcal{P}}+\text{Tr}\{\boldsymbol{\mathcal{L}}_{SB}U_0(s,t_0)\boldsymbol{\mathcal{P}}U_0(t_0,s')\boldsymbol{\mathcal{L}}_{SB}\}_B. \tag{15}$$

We are mainly interested in phase coherence times and relaxation rates. The memory kernel is sufficient to calculate these quantities.

Within the approximation, which we will use, the resulting rates are identical to rates obtained by Fermi's golden rule. However, the advantage of the Liouville formalism is that it provides us a general framework to obtain the response functions and the rates in which we also could go beyond the approximations equivalent to Fermi's golden rule.

## IV. A PAIR OF DEFECTS

The Hamiltonian of a pair of TLS's is given by

$$H_{\text{Pair}}=-\frac{\Delta_1}{2}\sigma_x^{(1)}-\frac{\Delta_2}{2}\sigma_x^{(2)}-J\sigma_z^{(1)}\sigma_z^{(2)}, \tag{16}$$

where $J$ is the coupling between the TLS's and the $\Delta_i$ are the energy splittings of the TLS's. The phonons couple via their distortion field $e_{\alpha\beta}(\mathbf{r})$ to the elastic moments $\gamma^{(i)}_{\alpha\beta}$ of the TLS's.[16] Since we have no detailed information about the elastic moments, we approximate this coupling by

$$H_{SB}=\sum_k \{\sigma_z^{(1)}\lambda_k^{(1)}(\mathbf{r}_1)+\sigma_z^{(2)}\lambda_k^{(2)}(\mathbf{r}_2)\}(b_k+b_{-k}^\dagger) \quad (17)$$

with

$$\lambda_k^{(i)}(\mathbf{r}_i)=\gamma^{(i)}\cos(\Theta_i)ike^{i\mathbf{kr}_i}\sqrt{\frac{\hbar}{8Nm\omega_k}}, \quad (18)$$

where we introduced the angle $\Theta_i$ between the elastic moment and the direction of propagation of the phonon with wave vector $\mathbf{k}$. Since TLS's are one dimensional, we cannot account for the full angular dependence of the quadrupole coupling. Simplifying to a picture of a dipole coupling, the elastic moment couples to the direction of propagation of the phonons leading to the term $\cos(\Theta_i)$ with the angle $\Theta_i$ between the two vectors. Since we integrate over all directions of propagation of the phonon, this angle dependence yields only a numerical factor for a single TLS.

In the case of a pair of TLS's the phonons couple to the total moment of both defects. If the wavelength of the phonon scattered by the pair of defects is longer than the distance between them, the damping rates depend on the relative angle between the two TLS's. For defects further apart the resulting damping rates are independent of the relative angle between the two TLS's and the above introduced angular dependence yields only a numerical factor in the damping rates like for single TLS.

The same argumentation holds true for the coupling between an external electric field and the electric dipole moment of the defects as seen in the following section for the Rabi frequencies of the pair. Usual measuring frequencies fall in the range between 1 kHz and 1 GHz which gives wavelengths not shorter than 1 cm. Accordingly the field is uniform over the entire sample. For describing the angular dependence of the electric dipole coupling we introduce the angles $\theta_i$ between the dipole moments and the electric field.

### A. A pair without coupling to phonons

Before we go into detail about the calculations in Liouville space, we discuss the Hamiltonian (16) of the pair following Thimmel *et al.*[17] The transformation matrix

$$T=\exp\left(i\frac{\alpha}{2}\sigma_y^{(1)}\sigma_z^{(2)}+i\frac{\beta}{2}\sigma_z^{(1)}\sigma_y^{(2)}\right), \quad (19)$$

with $\tan(\alpha+\beta)=-J/\Delta$, $\tan(\alpha-\beta)=-J/\delta$, $\Delta=(\Delta_1+\Delta_2)/2$, and $\delta=(\Delta_1-\Delta_2)/2$ yields

$$H_D=T^\dagger H_{Pair}T=-\frac{\epsilon_1}{2}\tau_x^{(1)}-\frac{\epsilon_2}{2}\tau_x^{(2)}, \quad (20)$$

with the energies $\epsilon_{1/2}=\sqrt{J^2+\Delta^2}\pm\sqrt{J^2+\delta^2}$ and the Pauli matrices $\tau_x$, $\tau_y$, and $\tau_z$. The same transformation has to be applied to the dipole operators. In Fig. 1 we show the spectrum of a pair with $\Delta=1$, $J=1/6$, and $\delta=0$. The spectrum consists of four levels, the second and third of which are split by $2\sqrt{J^2+\delta^2}$, which is independent of $\Delta$. The arrows in Fig. 1 indicate possible transitions that can be induced by phonons or electric fields. Even though the Hamiltonian Eq. (20) describes two independent effective TLS's, the transformation Eq. (19) complicates the coupling to phonons or fields. Each phonon can induce all four transitions shown in Fig. 1. Since the phonon couples to the total moment of both TLS's, the four transitions are induced with two different transition strengths indicated by the numbers next to the arrows.

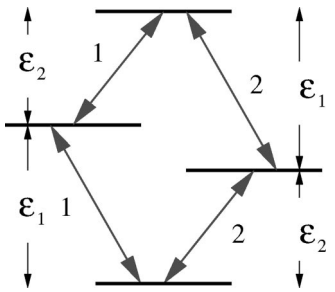


FIG. 1. The spectrum of a pair of weakly coupled TLS's. The arrows give the possible transitions that can be induced by phonons or electric fields.

The transformed dipole operators $\sigma_z$ are given by

$$T^\dagger\sigma_z^{(1)}T=u_1\tau_z^{(1)}+v_1\tau_x^{(1)}\tau_z^{(2)},$$

$$T^\dagger\sigma_z^{(2)}T=u_2\tau_z^{(2)}+v_2\tau_z^{(1)}\tau_x^{(2)},$$

with

$$u_1=\cos(\alpha), \quad v_2=-\sin(\beta),$$

$$u_2=\cos(\beta), \quad v_1=-\sin(\alpha).$$

In the case $J<\delta<\Delta$ the two systems remain independent since $v_i\sim O(J/\delta,J/\Delta)$ and $\epsilon_i\simeq\Delta_i+O(J/\delta,J/\Delta)$. Accordingly the coupling is negligible. This shows that the interaction can only affect *resonant* systems which means that both systems have the same energy splitting.

Below we focus on the case of $\delta<J\ll\Delta$ where $u_1\simeq -u_2\simeq -v_1\simeq -v_2\simeq 1/\sqrt{2}$. As long as the wavelength of the electromagnetic field is large compared to the distance of the two dipoles, the probabilities of the transitions in Fig. 1 induced by an electric field are proportional to the square of the transition strengths $s_\mp^2=(1/2)[\cos(\theta_1)\mp\cos(\theta_2)]^2$ [where $-$ $(+)$ corresponds to the transition marked by 1 (2)]. The $\theta_i$'s are the angles between the electric field and the dipoles of the defect $i$.

Assuming that the angles $\theta_i$ are evenly distributed, we obtain a broad distribution of transition strengths $s_\mp$. Nevertheless the modifications of the dielectric constant of the pair in comparison to isolated systems are of order $O(J/\Delta)$, and therefore small. In contrast, if one performs *phase-coherent* experiments on the tunneling defects, the situation changes. With rotary echos one is able to measure the Rabi frequencies of the pairs which depend on the transition

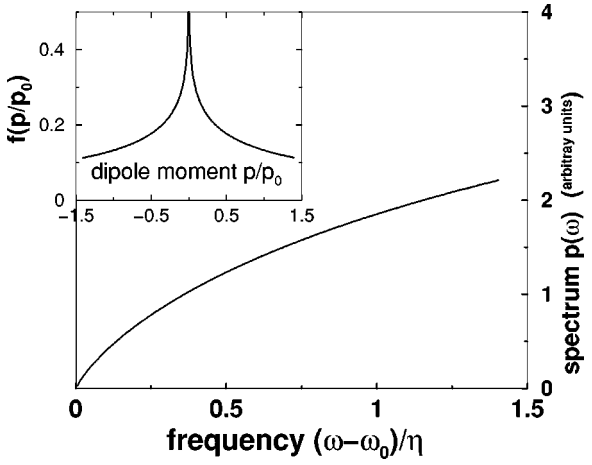


FIG. 2. The spectrum of the time-dependent dipole expectation value and the distribution of *effective* dipoles which results from mapping the Rabi frequencies of an ensemble of weakly coupled pairs onto an ensemble of isolated TLS's.

strengths. Thus, this quantity is broadly distributed in the case of weakly coupled pairs, whereas it should be peaked for identical isolated systems. Thimmel *et al.*[17] investigated a pair interacting with a quasiclassical electric field. The weak-coupling case is given as the third case ($n=m=1$) in their Appendix A4. The only additional assumption we need in order to adopt the results, is that the coupling $J$ is bigger than the field energy $\eta=pE$ using the bare dipole moment $p$ and the electric field $E=|\vec{E}|$. If $J$ is smaller, we would expect the coupling to be irrelevant compared to the field.

Taking only the dominant contributions into account, the time-dependent dipole expectation value of a pair excited by an external field with frequency $\omega_0$ reads

$$p^{(0)}(t)=\frac{1}{4}p_0\sum_{\pm}\, s_{\pm}r_1(1\pm r_2)\cdot\{\sin((\omega_0-\Omega_{\pm})t) - \sin((\omega_0+\Omega_{\pm})t)\} \tag{21}$$

with the Rabi frequencies $\Omega_{\pm}=\eta s_{\pm}$, the temperature-dependent factors $r_i=\tanh(\beta\epsilon_i/2)$, the frequency $\omega_0$ of the external field which is chosen to equal $\epsilon_1$ and the bare dipole moment $p_0$ of a single tunneling system. We assumed $J>0$. If $J<0$ the temperature-dependent factor would change to $r_1(1\mp r_2)$.

If we assume that we can describe many weakly coupled tunneling systems by an ensemble of weakly coupled pairs with different couplings,[18] the Rabi frequencies should be broadly distributed due to the distribution of the transition strengths $s_{i\mp}$. Figure 2 shows the spectrum of $p^{(0)}(t)$ after averaging over the angles $\theta_i$. We show only the part for frequencies higher than the external exciting frequency $\omega_0$. If we try to explain such a spectrum by an ensemble of noninteracting TLS's we have to assume the following distribution of dipole moments:

$$f(\tilde{p})=\frac{1}{2}\sum_{\pm}\int_0^{2\pi}\frac{d\theta_1}{2\pi}\int_0^{2\pi}\frac{d\theta_2}{2\pi}\delta(\tilde{p}-s_{\pm}) \tag{22}$$

$$=\frac{1}{2\sqrt{2}\pi^2}\int_{-1}^{1}dy\frac{\Theta(1-|\sqrt{2}\tilde{p}\pm y|)}{\sqrt{1-y^2}\sqrt{1-(\sqrt{2}\tilde{p}\pm y)^2}}, \tag{23}$$

with the Heaviside function $\Theta(x)$ and the normalized dipole moment $\tilde{p}:=p/p_0$. We, thereby, assumed equal probabilities for positive and negative coupling $J$, and we assumed weak coupling $\epsilon_1\simeq\epsilon_2\simeq\Delta$. The inner plot in Fig. 2 shows the distribution of dipole moments. The distribution is smooth between $\pm\sqrt{2}$ except for a logarithmic singularity for $p=0$.

We should point out once again that this distribution of *effective* dipole moments is only relevant for echo experiments since we extracted it from a distribution of Rabi frequencies mapping these to effective dipole moments in order to describe experiments within a picture of identical isolated tunneling systems. In the following section we take the coupling to phonons into account.

### B. Damped pair

In the last section we showed that as long as the tunneling systems are *resonant*, i.e., $\delta<J\ll\Delta$, even a weak coupling leads to a broad distribution of Rabi frequencies since the external wave is not able to resolve the single dipoles. We now investigate if something similar happens to the relaxation rates.

In order to obtain the damping rates, we have to calculate actual measuring quantities like the dipole susceptibility or the time-dependent dipole expectation value (starting from an off-equilibrium situation). Thus, we need the effective time evolution [cp. Eq. (13)] of the system and, thereby, the memory kernel [cp. Eq. (14)]. It is convenient to use the operators[19] $|ij)=|i\rangle\langle j|$, where the $|i\rangle$ are the eigenvectors of the pair Hamiltonian $H_{\rm Pair}$, as basis for the Liouville space. In this basis the free time evolution of the pair is the diagonal matrix

$$[\mathcal{U}_S^{-1}(z)]_{(ij)(kl)}=(ij|\mathcal{U}_S^{-1}(z)|kl)=-(z-(E_i-E_j))\,\delta_{(ij)(kl)}, \tag{24}$$

where the eigenvalues $E_i$ of the Hamiltonian $H_{\rm Pair}$ correspond to the eigenvectors $|i\rangle$.

#### 1. Bath functions

Before we discuss the memory kernel we introduce the relevant bath functions. We distinguish between correlations

$$B_C^{(ij)}(z)=\int_0^{\Omega_c}d\omega J^{(ij)}(\omega)\coth(\beta\omega/2)\,\frac{z}{\omega^2-z^2}, \tag{25}$$

and response functions

$$B_R^{(ij)}(z)=\int_0^{\Omega_c}d\omega 2J^{(ij)}(\omega)\frac{\omega}{\omega^2-z^2}, \tag{26}$$

with the generalized spectra $J^{(ij)}(\omega)$ [compare Eq. (10)] defined as

$$J^{(ij)}(\omega)=\frac{\pi}{2}\sum_k\ [\lambda_k^{(i)}(\mathbf{r}_i)]^\dagger\lambda_k^{(j)}(\mathbf{r}_j)\,\delta(\omega-\omega_k).$$

$J^{(ii)}(\omega)$ yield the well-known spectral functions whereas the mixed spectra $J^{(i\neq j)}(\omega)$ show an explicit dependence on the positions of the TLSs $i$ and $j$:

$$[\lambda_k^{(i)}(\mathbf{r}_i)]^\dagger\lambda_k^{(j)}(\mathbf{r}_j)\propto\exp[i\mathbf{k}(\mathbf{r}_j-\mathbf{r}_i)]. \tag{27}$$

But, as long as $[\mathbf{k}(\mathbf{r}_j-\mathbf{r}_i)]\ll 1$, which means as long as the wavelength of the phonon is larger than the distance between the TLSs, the mixed spectra are equal to the spectral functions. In order to calculate the spectra explicitly, one assumes linear dispersion and one introduces an upper cut-off $\Omega_c$ for the phonon frequencies.[12]

#### *2. Approximations*

It is well known that if you start with two arbitrary TLS's coupled to a boson bath, the coupling generates an effective interaction between the TLS's through exchange of virtual bosons.[20] A careful investigation shows that the entries in the memory kernel proportional to the bath response functions $B_R^{(i\neq j)}(z)$ provide this effective interaction. We discuss the interaction between the TLS's as a renormalized coupling which incorporates these effects already. Therefore we neglect the contributions from $B_R^{(i\neq j)}(z)$ in the memory kernel.

In the weak-coupling limit the resulting memory kernel yields only a small correction to the frequencies of the free dynamics. Representing the memory kernel and the free time evolution [cp. Eq. (13)] as a matrix in the basis given above, we neglect all entries of the memory kernel which combine different invariant subspaces of the free time evolution. This is valid if the coupling $J$ is bigger than typical entries of the memory kernel. Otherwise, the coupling would turn out to be completely irrelevant, resulting in two uncoupled TLS's. Additionally, we evaluate the remaining entries of $M(z)$ at the resonance frequency of the corresponding invariant subspace of the free time evolution.[14]

The real parts of the memory kernel yield small renormalizations for the resonance frequencies of our system. We are interested in the dephasing and relaxation rates which are given by the imaginary parts.

#### *3. Decoherence rates*

If we are interested in the dipole susceptibility, we have to calculate the time evolution of the subspaces involving the energies $\pm\epsilon_1$ and $\pm\epsilon_2$. The four subspaces are independent from each other and within our weak-coupling assumption their dynamics are equivalent. We obtain for the memory kernel of the subspace of frequency $-\epsilon_1$

$$M(-\epsilon_1)=\begin{pmatrix} A+B_1-C & -B_2 \\ -B_1 & A+B_2+C \end{pmatrix} \tag{28}$$

with

$$A=[\cos^2(\Theta_1)+\cos^2(\Theta_2)]B_C^{(ii)}(-\epsilon_1),$$

$$C=\cos(\Theta_1)\cos(\Theta_2)[B_C^{(12)}(-\epsilon_1)+B_C^{(21)}(-\epsilon_1)],$$

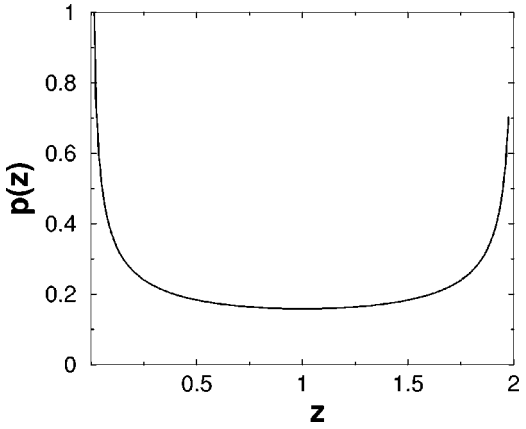


FIG. 3. The distribution of the factor $z$ entering multiplicative into the rate $\Gamma_1$.

$$B_{1/2}=2[\cos^2(\Theta_1)+\cos^2(\Theta_2)]\cdot[B''^{(ii)}_C(-\epsilon_2) \mp B''^{(ii)}_R(-\epsilon_2)],$$

where $B''^{(ii)}_{C(R)}$ is the imaginary part of the bath correlation (response) function.

A direct way of observing these rates is to perform two-pulse echo experiments. Therein the decay of the echo amplitude is governed by the rates we obtain from the above memory kernel.

There are two simple extreme cases:

(a) $\lambda>r_{12}$: If the wavelength $\lambda=hc/\epsilon_1$ is larger than the distance $r_{12}$ between the two TLSs, the generalized spectra are independent of the positions of the TLS's. Since the real parts of the bath functions are of order $O(\alpha\Omega_c^2)$, whereas the imaginary parts are of the order $O(\alpha\Delta^2)$, the off-diagonal elements in $M(-\epsilon_1)$ are negligible. The angles $\Theta_i$ are not independent of each other since the relative angle $\psi_{12}$ between the two TLS's is fixed for each configuration. The average over all possible directions of the phonon yields $\overline{\cos(\Theta_1)\cos(\Theta_2)}=(1/4\pi)\cos(\psi_{12})$, so that the rates depend on the relative angle between the two TLSs.

We obtain two transversal rates for excitations with energy $\epsilon_1$ corresponding to the transition from the ground state to the first excited state ($\Gamma_1$) and from the first to the second excited state ($\Gamma_2$). The expression $A$ in $M(-\epsilon_1)$ yields the part already present for an isolated system. The expressions $B_{1/2}$ reflect the loss of coherence if the pair undergoes a thermal fluctuation with energy $\epsilon_2$. The term $C$ describes the mixing since the phonon cannot resolve both dipoles independently. Finally we get

$$\Gamma_1=\Gamma_0[1-\cos(\psi_{12})][1+2n(\epsilon_1)]+2\Gamma_0 2n(\epsilon_2),$$

$$\Gamma_2=\Gamma_0[1+\cos(\psi_{12})][1+2n(\epsilon_1)]+2\Gamma_0[1+2n(\epsilon_2)], \tag{29}$$

with the bose factors $n(\epsilon_{1/2})$ and a material constant $\Gamma_0$. For an ensemble of pairs with homogeneously distributed relative angles $\psi_{12}$ we end up with a broad distribution of phase decoherence rates. In Fig. 3 we show the distribution of the

angle dependent factor $z := 1-\cos(\psi_{12})$. In the case of an isolated TLS, $z=1$.

The distribution for the factor $z$ follows

$$p(z) = \frac{(1/2\pi)}{\sqrt{2z-z^2}} \quad \text{with} \quad z \in ]0,2[. \tag{30}$$

It shows two square-root singularities. Since for $T<\Delta$ the bose factor $n(\epsilon_{1/2}) \ll 1$, the minimal rate $\Gamma_1^{\min}(z=0) = 4\Gamma_0 n(\epsilon_2)$ is very small leading to long coherence times.

(b) $\lambda < r_{12}$: In the case of $\lambda < r_{12}$ the terms $C$ are negligible. In this case the dynamics turns out to be identical to that of two isolated TLS's with energies $\epsilon_i$ which are independently coupled to a heat bath, and the rates are not distributed.

So far, we only discussed the decoherence rates. A numerical analysis of the relaxation rates, determining the decay into thermal equilibrium, shows that these are also broadly distributed due to the same mechanism.

### C. [100]- and [111]-tunneling defects

In this section we try to give a short overview (without the lengthy calculation) what we expect for tunneling defects taking their full structure into account. We assumed, so far, that the dielectric dipole as the elastic moment of a tunneling defect can point in any direction and both are not necessarily aligned. Accordingly all four angles $\Theta_1$, $\Theta_2$, $\theta_1$, and $\theta_2$ are independently distributed. These assumptions seem natural for tunneling defects of which we do not know the structure. The structure of tunneling defects in crystals, however, is usually well known. There are three possible cases for cubic host crystals: 6, 8, or 12 potential minima which represent the face centers, the corners and the edge centers of a cube. In the case of six (eight) minima the dipole moment points along a [100] direction ([111] direction). These defects are, therefore, called [100] defects ([111] defects).

For an electric field in $x$ direction $\mathbf{E}=[E,0,0]$ the angle between the electric field and the dipole moment can only take the values: 0, 90, and 180 deg for a [100] defect and 45 and 135 deg for a [111] defect. Calculating the time-dependent dipole expectation value for a rotary echo experiment [cp. Eq. (21)], we end up with more different transition strengths $s_i$ than the two $s_\pm$ of the case of two-level systems. For a pair of [100] defects we would obtain $s_0=0$, $s_1=1$, and $s_2=2$ leading to two visible peaks in the Rabi-frequency spectrum [the peak corresponding to $s_0$ has vanishing amplitude, comp. Eq. (21)]. In the case of a pair of [111] defects we obtain $s_0=0$ and $s_1=\sqrt{2}$ and, thus, only one peak in the Rabi-frequency spectrum. The situation turns around for measuring with an electric field in [111] direction where the [111] defect exhibits two Rabi frequencies and the [100] defect only one.

Using the full structure of the tunneling defects we can take into account the tensor character of the elastic moment (see Refs. 3,10). The assumption that the electric and the elastic moment are independently directed ensured within the two-state approximation that the dielectric susceptibility of pairs with a dephasing rate $\Gamma_1(\psi_{12}=0)$ [cp. Eq. (29)] was finite (due to a finite total electric moment). The dielectric response of a pair of [111] defects by measuring in $x$ direction exhibits a contribution decaying with a rate $\Gamma_1(\psi_{12}=0)$ which has a finite total electric moment despite the fact that the moments are not independent of each other. Therefore, our assumption of independent directed moments is necessary within the two-state approximation to reveal this situation. Instead of a broad distribution of rates we get for this special case discrete values which we can describe with a set of discrete prefactors $z=0,1,2$.

The dynamics of a pair of tunneling defects taking their full structure into account depends strongly on the geometry of the experiment. Typically, we have several Rabi frequencies and we have long dephasing times $\Gamma_1^{-1}(\psi_{12}=0)$. For a detailed comparison with experiments a complete investigation would be necessary. However, assuming independent homogeneous distributions of the angles between the elastic moment and the elastic waves and the dielectric dipole moment and the electric field for both tunneling systems forming a pair reveals the full range of possible Rabi frequencies and damping rates for tunneling defects in crystals.

## V. MANY TWO-LEVEL SYSTEMS

In the last section we described weakly coupled pairs of tunneling defects. In this section we argue that we can describe weakly coupled tunneling defects as an ensemble of pairs; so to say within a pairmodel.

In a crystalline host material like KCl doped with a few ppm of defects, for example 5 ppm of $^6$Li, these defects form tunneling systems which tunnel between various minima of their potential-energy landscape provided by the host. Measurements of the dielectric constant, for example, are well described within a picture of isolated identical tunneling defects with a tunneling splitting of about $\Delta \simeq 1.65$ K.[23] At concentrations of about 60 ppm the interaction between the tunneling defects results in measurable deviations from the picture of isolated identical tunneling defects.[23] At these medium concentrations the *pairmodel* introduced by Klein and others[24] is used to describe the systems. Within this approach one describes the tunneling defects as an ensemble of pairs of two-level systems. At higher concentrations of defects the pairmodel fails and a more elaborated treatment of the coupling is necessary.[23] A model by Würger describes most experiments very well.[23,3]

A delocalization of the excitations of the tunneling defects or in other words the existence of a collective phase of tunneling defects is experimentally not found and at concentrations below 10 ppm the interaction between the tunneling defects can be discarded at all for describing data of the dielectric constant. We showed in the last section by using a pairmodel approach that even very weak couplings lead to several values of Rabi frequencies and relaxation rates instead of single values expected in a picture of isolated identical tunneling defects. These effects might help to understand the small deviations of the experiments at samples with dopings less than 10 ppm and the theoretical predictions. Finally, it leads to a clearer understanding of a single tunneling defect in these crystalline materials.

In the following we will underpin the use of the pair-model. If we neglect the coupling to phonons the Hamiltonian (9) is identical to an Ising spin-$\frac{1}{2}$ system with a transversal field $\Delta$. The coupling $J_{ij}$ between the spins depends on the inverse of the distance $r_{ij}$ between two spins cubed: $J_{ij} \propto 1/r_{ij}^3$. Due to the angle dependence of the dipole-dipole type interaction the coupling constant $J_{ij}$ can have both signs. If we focus our consideration for a moment to a single TLS $i$ we can define the local fields $h_j = J_{ij}\sigma_{zj}$ which a neighbor $j$ is providing by the interaction to the TLS $i$. The total local field $h = \Sigma_j h_j$ vanishes due to the symmetry $P(-J) = P(J)$ of the distribution $P(J)$ of coupling constants $J_{ij}$. However, in order to decide whether the interaction is a relevant quantity for the dynamics of the TLS $i$ we might sum up the absolute values: $w = \Sigma_j |h_j|$. Switching over from a sum to an integral in real space and integrating only over all neighbors within a sphere of radius $R$ yields

$$w = \int_{R_{min}}^{R} dR 4\pi R^2 \frac{J_0}{R^3} \tanh(\beta\Delta/2)$$

$$= 4\pi J_0 \tanh(\beta\Delta/2) \ln\left(\frac{R}{R_{min}}\right), \tag{31}$$

with a minimal distance $R_{min}$ between TLS's given by the lattice constant and a mean interaction $J_0$ between TLSs. In the thermodynamic limit $R \to \infty$ this expression diverges for any strength of the coupling. Accordingly, we expect that the interaction dominates the dynamics of the system. Let us try to base the above arguments on a more rigorous footing. The problem is the diagonalization of the Hamiltonian

$$H = \sum_i \frac{\Delta}{2}\sigma_{zi} + \sum_{ij} J_{ij}\sigma_{xi}\sigma_{xj},$$

where we rotated the Pauli matrix basis around $\sigma_y$ by 90 deg compared to Eq. (9). Without interaction between the TLSs, the ground state would be given by the product state of all the ground states of the individual TLS's. The first excited state would be $N$-fold degenerated (with the number $N$ of TLS's). The wave functions are the products of $N-1$ of the TLS's in their ground state and one TLS in its excited state. Tuning on weak interactions we might be tempted to neglect intermixture of the ground, the first excited and higher excited states and to discuss only the lifting of the degeneracy of, for example, the first excited state. Its Hamiltonian is given by

$$W_1 = \begin{pmatrix} 0 & J_{12} & J_{13} & \cdots \\ J_{12} & 0 & J_{23} & \cdots \\ J_{13} & J_{23} & 0 & \cdots \\ \vdots & \vdots & \ddots & \end{pmatrix}, \tag{32}$$

with the coupling $J_{ij}$ between TLS $i$ and TLS $j$ where we used the basis: $(\uparrow\downarrow\downarrow\cdots)$, $(\downarrow\uparrow\downarrow\cdots)$, .... The eigenvalues of the matrix $W_1$ are distributed between $[-v,v]$ with $v = v_j = \Sigma_j |J_{ij}|$ which is independent of $j$. Since $w = v \tanh(\beta\Delta/2)$ we know from the considerations above that in the thermodynamic limit $v$ diverges. Accordingly, the assumption that we can neglect intermixture of the ground, the first excited and higher excited states is questionable. We expect that the excitations are delocalized and the system is in a collective phase. Levitov[21] and Parshin and Schober[22] found in similar models such a delocalization for excitations interacting with a dipolelike long-range $1/r^3$ interaction.

These theoretical results are in contrast to experimental results. In a typical experimental situation the sample size is of the order of 1 mm$^3$. With lattice constants of the order of 5 Å and assuming $T=0$ we can estimate that only for mean couplings $J_0 \geqslant \Delta/[4\pi \ln(1\text{ mm}/5\text{ Å})] \simeq 5\times 10^{-3}\Delta$, the interaction actually dominates the dynamics of the tunneling defects. Accordingly, at lowest concentrations (resulting in lowest mean couplings) the experimental results might not contradict the consideration above. However, we neglected in our theoretical investigation the coupling to phonons (besides the possibility that this coupling gives rise to the coupling between the TLS's). The coupling to phonons results in a finite linewidth $\gamma$ for the excitation frequencies of the TLS's.[27] If an excited TLS is coupled by an interaction $J$ to a neighbor the excitation energy is transferred back and forth between the TLS and its neighbor with a period $2\pi/J$. If the lifetime (relaxation time) $\tau = 1/\gamma$ of the excitation of the TLS is less than the period of the oscillation, the TLS would be decayed to its ground state before the energy could be transferred to the neighbor. The coupling would be irrelevant in this case since it is not effective in delocalizing the energy over both systems. Thus, the interaction with phonons yields a natural cut-off radius $R_c$ for the interaction by the argument $J(R_c) = J_{max}(R_{min}/R_c)^3 = \gamma$ with the minimal distance $R_{min}$ between tunneling defects which is of the order of a lattice constant and the maximal coupling $J_{max}$ between nearest neighbors. Using this cut-off radius in the expression above for estimating the relevance of the interaction yields $w' = 4\pi J_0 \tanh(\beta\Delta/2)\ln(R_c/R_{min})$ since we only have to integrate over the neighbors within a sphere of radius $R_c$. Following our simple argumentation we would conclude that for $w' < \Delta$ the interaction between the TLSs does not lead to a collective behavior/ delocalization. This argument does not rule out the formation of pairs or clusters of more than two TLSs but it explains the absence of a delocalization of the single TLS excitations. The same argument holds in glasses.[7]

To quantify the influence of a neighbor which is further apart form the TLS of interest than the cut-off radius we investigate a Gedanken experiment. We prepare a symmetric TLS in a superposition between its ground and excited state. This superposition is actually a localized state (in real space) with a finite dipole expectation value. The free evolution forces the tunneling particle to tunnel periodically between its two potential minima with a period $2\pi/\Delta$ where $\Delta$ is the tunneling element. The coupling to phonons leads to a finite lifetime for the coherent superposition resulting in an average behavior for the dipole expectation value of $p(t) = \cos(\Delta t)\exp(-\gamma t)$ with time $t$. If we include one neighbor with a coupling $J$ to the TLS we can simply calculate the time dependence of the dipole expectation value with the methods described in the section before. We assume that the coupling is *irrelevant*: $J < \gamma$. To simplify the investigation we assume furthermore that the distance $r_{12}$ between the two

TLS's is bigger than the wavelength $\lambda$ of a phonon in resonance with the TLS's. This saves us from taking additionally mixed bath functions into account which would complicate the analysis, but which would not change the general behavior. Under these assumptions the corrections to the dipole expectation value are of order $O((Jt)^2)$. The assumption $J < \gamma$ guarantees that the correction given by one neighbor is small for all times. However, it is important for the case of many neighbors that the corrections given by a neighbor are quadratic. All neighbors whose distance to the TLS of interest is bigger than the cut-off radius give a correction $\propto \Sigma_j J_{ij}^2 t^2 \simeq [J_0 J(R_c)] t^2$ where $j$ sums only over neighbors with $r_{ij} > R_c$. For $J_0 > J(R_c)$ it holds that $\sqrt{J_0 J(R_c)} > \gamma$ and the corrections are visible. However, it also ensures that the correction of nearby neighbors $r_{ij} < R_c$ is bigger than the correction by all the distant neighbors, which allows us to neglect the distant neighbors in a first approach completely. Since typically $J_0, J(R_c) \ll \Delta$ all corrections to the dynamics of the defects are small and we do not expect a collective phase to be formed or a delocalization of the excitations to happen.

At this point we will investigate what are typically cut-off radii and how many neighbors will be within a sphere of the cut-off radius around a given defect. The linewidth of $CN^-$ defects in KCl, for example, is about $\gamma \simeq 1.35 \times 10^9\ s^{-1}\hbar \simeq 10\ \mathrm{mK} k_B$.[10] Typical nearest-neighbor maximal couplings[3] are between $10^2$–$10^4$ K with typical lattice constants of about 0.5 nm ($a_{\mathrm{KCl}} \simeq 0.623$ nm at 1 K). Thus, the cut-off radius is a few hundreds times the lattice constant. The number of defects $\#n$ within a sphere of radius $R_c$ can be estimated as $\#n = c(R_c/R_{min})^3 \simeq c \times 10^4$–$10^6$ with the concentration $c$ of defects. For concentrations of a few ppm we expect the number of neighbors within reach of the coupling to be of order $O(1)$ or less.

So far, we argued that the interaction between the TLSs does not lead to a delocalization of the excitations since the coupling to phonons provides an effective cut-off radius for the otherwise long-range $1/r^3$ interaction. The same argument was used by Burin and Kagan[7] to conclude that there is no delocalization of the excitations of the single tunneling systems in glasses.

However, Burin and Kagan further showed that in glasses the weak coupling between the tunneling systems leads to a formation of resonant pairs. These resonant pairs can be mapped to new effective TLS's. If the tunneling systems have finite asymmetry energies these effective TLS's are coupled to each other as shown in the Appendix. Since the coupling between the resonant pairs is of the same order of magnitude than the coupling leading to the resonant pairs these effective TLSs are strongly coupled to each other and the excitations delocalize. As Burin and Kagan showed this delocalization is accompanied by spectral diffusion which also arises only for asymmetric tunneling systems.[25] Thus, the formation of a collective phase due to the interaction of resonant pairs competes with the destruction by spectral diffusion and from this competition a new mechanism for energy relaxation emerges.

Resonant pairs formed from symmetric TLS's, however, do not interact with each other by direct virtual phonon exchange. Nevertheless the resonant pairs interact with each other by higher-order effects (in the coupling to phonons), i.e., two-phonon exchange. This coupling is weaker and the resonant pairs are only weakly coupled. The same arguments, which lead us to conclude that the single TLS's excitations do not delocalize, therefore forbid a delocalization of the resonant pairs. What is more, the higher-order coupling depends typically on the distance $r$ between the TLS's like $1/r^n$ with $n > 3$. Thus, the coupling is of short-range nature and in a short-range coupling problem we only expect collective phases for couplings comparable to the energy splittings of the effective pairs.

After all, we believe to have ruled out the occurrence of a collective phase. The main argument is that as long as the coupling is weaker than the linewidth of an excitation the coupling can be neglected. Any progress in a full and comprehensive understanding of the above given argument would be desirable. Accepting this argument to hold true, let us conclude that the interaction between tunneling defects in mixed crystals does not lead to a collective behavior at least as long as the coupling is sufficiently weak. Nevertheless, the defects will form pairs, triples and clusters of more defects. In this scenario of small clusters the pair will obviously be the most important member since more than two defects with equal couplings between each other are more unlikely than simply two such defects. In a triple, where two of the three are stronger coupled to each other than both are to the third partner the pair is only weakly affected by the third partner, and we simply disregard these corrections in our approach. It would be interesting to take triples and bigger clusters equally into account, and this step would be necessary to obtain a quantitative understanding of experiments. However, to take only pairs into account seems to capture the main features which was excluded so far from any consideration: Even at very small defect concentrations the interaction effects cannot be neglected since the system forms small clusters (mainly pairs), which leads to very different Rabi frequencies and relaxation and dephasing rates compared to the case of isolated identical tunneling defects.

Below we briefly discuss the experimental relevance of our results.

## VI. EXPERIMENTAL RELEVANCE

The low-temperature properties of defect crystals are studied in thermal, acoustic, and dielectric experiments, which measure *bulk* quantities such as specific heat, thermal conductivity, speed of sound, internal friction, dielectric constant, and dielectric loss. Furthermore there are various dielectric echo experiments measuring directly the relaxation rates or the Rabi frequencies. As we have shown in the previous sections, small interactions between the tunneling defects yield broad distributions of effective moments within the two-state approximation. Taking the full structure of the tunneling defects into account, we obtain a set of discrete values for the effective moments. This leads to several different Rabi frequencies and relaxation or phase decoherence rates, but all the *bulk* quantities mentioned above are hardly affected.

Typical tunneling elements vary between $\Delta \simeq 2.3$ K for NaCl:$OD^-$ and $\Delta \simeq 85$ mK for NaF:$OH^-$.[26] Dielectric experiments are usually performed at temperatures that coincide with the tunneling elements which means between $T \simeq 10$ mK and a few Kelvin. Typical measuring frequencies range between a few hundred hertz and about 1 GHz corresponding to energy splittings between 50 nK and 50 mK. These frequencies are too small to excite single tunneling systems or weakly coupled pairs resonantly except for the NaF:$OH^-$ systems.

The easiest way to confirm our results would be to perform echo experiments. Therein the relaxation rates and dephasing rates of the defects, which are in resonance with the external field, are obtained from the decay of the echo amplitude. However, experiments in NaF doped with a few ppm of $OH^-$ showed that the energy splittings of the tunneling defects are broadly distributed.[26] One does not know, if the sample is heavily strained or if the interaction between the defects is strong. In both cases our approach is not adequate.

The system NaCl:$OH^-$ with a tunneling splitting of about $\Delta_0 \simeq 1.6$ K has been well studied. At an OH concentration of about 16 ppm, Ludwig[26] measured the dielectric loss and constant. They report a relaxational contribution in the dielectric constant which also leads to a measurable dielectric loss. The relaxational contribution occurs due to internal strains, which also lead to a distribution of energy splittings and thereby to a distribution of relaxation rates. The data of the dielectric loss shows that the relaxation rates are distributed over six orders of magnitude. Describing the tunneling defects as two-level systems and neglecting interactions between them the relaxation rates depend on the asymmetry $F$ resulting from the strain: $\Gamma \propto \Delta_i^2 \sqrt{\Delta_i^2 + F^2}$.[27] The distribution of asymmetries necessary to describe the data is unrealistic broad. If we take the full structure of the NaCl:$OH^-$ system as a [100] defect into account but we still neglect interactions between the defects, the dependence of the relaxation rates on the various asymmetries changes. Nevertheless, taking only a distribution of internal strains (asymmetries) into account in order to explain the dielectric loss actually disagrees with the data of the dielectric constant.[28] By including weak interactions between the tunneling defects, we showed that we obtain a set of considerably different rates. If the asymmetries broaden the discrete peaks in the rate distribution in a way as discussed above, this leads naturally to very broad distributions. Thus, the relaxational contributions in the dielectric loss and the dielectric constant results from internal strains but the broad distribution of relaxation rates necessary to describe the data results from the interplay of weak interactions between the tunneling defects and asymmetries. Accordingly, the data discussed yields a strong but indirect hint to the experimental relevance of our investigation.

Similar broad distributions of rates were found in KCL-crystals doped with 2 ppm $OH^-$.[26] The tunneling splitting in these systems is about $\Delta \simeq 170$ mK. Accordingly, this system is an ideal candidate for echo experiments for two reasons. First, the resonance frequency of about 4 GHz is technically accessible. Second, because of the dielectric constant data, one would suggest that the tunneling systems are only weakly disturbed by strain fields which simplifies the interpretation of the results considerably. Therefore, we strongly recommend echo experiments in these materials, since it would be the first direct observation of the dynamics of well-defined tunneling systems in crystals.

## VII. SUMMARY

We have investigated the influence of weak (transversal) interactions between impurities in alkali halides. Here is a brief summary of the main points:

(i) Since the coupling constants between the tunneling systems are broadly distributed and since the coupling to phonons leads to a finite lifetime for any excitation, we can describe the system as an ensemble of weakly coupled pairs. Thus, weak interactions do not lead to a relaxation mechanism via energy exchange.[29]

(ii) Weak couplings between the tunneling systems are only relevant as long as the systems are *resonant*—the difference of the two energy splittings is smaller than the coupling $\delta = |\Delta_1 - \Delta_2| < J$ and as long as the inverse lifetime $\gamma_i$ of the eigenstate $|i\rangle$ is smaller than the coupling, $\gamma_i < J$.

(iii) Typical electric fields used to investigate the dynamics of tunneling impurities in alkali halides have wavelengths exceeding the mean distance of the impurities even at concentrations of only a few ppm. Accordingly the field cannot resolve the single dipoles but only the total moment. In a weakly coupled pair bulk quantities like the dielectric constant are hardly affected, but the pair exhibits two Rabi frequencies determined by the sum and the difference of the dipoles. If the relative angle between the dipoles of two tunneling systems are homogeneously distributed, the Rabi frequencies become broadly distributed between zero and twice the Rabi frequency of an isolated impurity. For tunneling defects in crystals the dipoles have discrete directions leading to a discrete set of Rabi frequencies.

(iv) Similar arguments hold true for the phase coherence times and the relaxation rates which are determined by the coupling of the phonons to the elastic moments. This leads to a discrete set of rates with a very small minimal rate compared to the rate of isolated impurities.

(v) Dielectric loss data in NaCl: 16 ppm OH show broadly distributed relaxation rates which cannot only be attributed to strain fields. These features can be explained within our weak-coupling picture. KCl samples doped with 2 ppm $OH^-$ show similar behavior, and we strongly recommend echo experiments in these materials since it would be a direct observation of the dynamics of well-defined tunneling systems in crystals.

## ACKNOWLEDGMENTS

The author wishes to thank H. Horner, S. Ludwig, B. Thimmel, and K. McRobert for many stimulating and clarifying discussions. Parts of the work were done in the Institut für Theoretische Physik at the University of Heidelberg. In addition I would like to thank the DFG, which supported the work in Heidelberg within the project HO 766/5-3 ''Wechselwirkende Tunnelsysteme in Gläsern und Kristallen bei

tiefen Temperaturen.'' The work in Stanford was supported by the Alexander-von-Humboldt foundation and the DOE Grant No. DE-FG03-90ER45435-A011.

## APPENDIX: INTERACTION BETWEEN RESONANT EFFECTIVE PAIRS

The main goal of this appendix is to show that resonant pairs formed by weakly coupled two-level systems can only interact with each other by the same interaction leading to the formation of the resonant pairs if the two-level systems are asymmetric. Therefore, we discuss four coupled TLS of which TLS 1 and 2 form the resonant pair $a$ and TLS 3 and 4 form the resonant pair $b$.

The Hamiltonian for these four two-state systems is given by

$$H=\sum_{i=1}^{4}\left(\frac{\Delta_i}{2}\sigma_x^{(i)}+\frac{F_i}{2}\sigma_z^{(i)}\right)+\sum_{i,j=1}^{4}J_{ij}\sigma_z^{(i)}\sigma_z^{(j)}, \quad \text{(A1)}$$

with the tunneling splittings $\Delta_i$, the asymmetries $F_i$ and the coupling between the two-state systems $J_{ij}$. Diagonalizing each two-state Hamiltonian independently results in

$$H=\sum_{i=1}^{4}\frac{\epsilon_i}{2}\sigma_z^{(i)}+\sum_{i,j=1}^{4}J_{ij}\{\bar{u}_i\bar{u}_j\sigma_z^{(i)}\sigma_z^{(j)}+u_iu_j\sigma_x^{(i)}\sigma_x^{(j)}\}$$
$$+\sum_{i,j=1}^{4}J_{ij}\{\bar{u}_iu_j\sigma_z^{(i)}\sigma_x^{(j)}+u_i\bar{u}_j\sigma_x^{(i)}\sigma_z^{(j)}\},$$

with the energy splittings $\epsilon_i$ and $\bar{u}_i=F_i/\epsilon_i$ and $u_i=\Delta_i/\epsilon_i$. We assume that $|\epsilon_1-\epsilon_2|\leqslant u_1u_2J_{12}$ and $|\epsilon_3-\epsilon_4|\leqslant u_3u_4J_{34}$. Thus, system 1 and 2 form a resonant pair $a$ and system 3 and 4 form a resonant pair $b$. It is the coupling term $u_1u_2J_{12}$ and $u_3u_4J_{34}$ which shifts the degeneracy of the up-down states $|\downarrow_1\uparrow_2\rangle=:|a,1\rangle$ and $|\uparrow_1\downarrow_2\rangle=:|a,2\rangle$ for the resonant pair $a$ and $|\downarrow_3\uparrow_4\rangle=:|b,1\rangle$ and $|\uparrow_3\downarrow_4\rangle=:|b,2\rangle$ for the resonant pair $b$. These terms turn out to be the effective tunneling splittings for this up-down effective two-state system: $\Delta_{pa}:=u_1u_2J_{12}$ and $\Delta_{pb}:=u_3u_4J_{34}$.

The effective two-state resonant pairs only consider the states $|a,1\rangle$ and $|a,2\rangle$ for the resonant pair $a$ and $|b,1\rangle$ and $|b,2\rangle$ for the resonant pair $b$, respectively. The effective Hamiltonian for these four states writes

$$H=\frac{\Delta_{pa}}{2}\tau_x^{(a)}+\frac{F_{pa}}{2}\tau_z^{(a)}+\frac{\Delta_{pb}}{2}\tau_x^{(b)}+\frac{F_{pb}}{2}\tau_z^{(b)}+J_{ab}\tau_z^{(a)}\tau_z^{(b)} \quad \text{(A2)}$$

with $F_{pa}=|\epsilon_1-\epsilon_2|/2$, $F_{pb}=|\epsilon_3-\epsilon_4|/2$ and

$$J_{ab}=\bar{u}_1\bar{u}_3J_{13}+\bar{u}_2\bar{u}_4J_{24}-\bar{u}_1\bar{u}_4J_{14}-\bar{u}_2\bar{u}_3J_{23}, \quad \text{(A3)}$$

and

$$\tau_x^{(a)}:=|a,1\rangle\langle a,2|+|a,2\rangle\langle a,1|,$$

$$\tau_z^{(a)}:=|a,1\rangle\langle a,1|-|a,2\rangle\langle a,2|.$$

The operators $\tau_x^{(b)}$ and $\tau_z^{(b)}$ are defined in the same way.

These results show that the effective resonant pairs, which Burin and Kagan[7] introduced, are indeed coupled with each other by the same interaction which leads originally to the formation of the resonant pairs if and only if the two-level systems are asymmetric. The interaction between resonant pairs is as strong as the parameters of the resonant pairs: $\Delta_{0pa}:=u_1u_2J_{12}\sim J_{ab}$. Thus, the resonant pairs are strongly interacting and they form a collective cluster. However, the result shows that the interaction between the resonant pairs vanishes for symmetric two-state systems.


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