Low temperature universality in disordered solids

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Abstract

The vast majority of solids in Nature are strongly disordered or amorphous. Many of these materials show "universality" in their low-temperature elastic properties, despite enormous differences in their microscopic structure\textsuperscript{1,2}. Discovered 40 years ago\textsuperscript{3}, this universality has emerged as one of the outstanding unsolved mysteries in condensed matter physics\textsuperscript{4}. Here we show that the mere presence of strong disorder and tunneling states symmetric under spatial inversion necessarily gives universality. Such tunneling states interact weakly with phonons, yet gap the non-symmetric tunneling states above a crossover energy. Thus neither glassiness nor any amorphous structure are required for universality, only strong disorder - a result concordant with recent experiments\textsuperscript{2}. 
Some features of the universality are well known. Thus, for a large variety of disordered and amorphous solids, for temperature $T < T_U \sim 3K$, the specific heat $C_v(T) \propto T^\alpha$ with $\alpha \approx 1$, the thermal conductivity $\kappa(T) \propto T^3$ with $\beta \approx 2$, and the internal friction or "quality factor" $Q(T) = 2\pi l(T)/\lambda(T)$, dividing the phonon mean free path $l(T)$ by the phonon thermal wavelength $\lambda(T)$, goes to a constant "plateau" value $Q_o \sim 10^3$. Both $T_U$ and $Q_o$ vary somewhat between materials (by up to $2-3$ on either side of the quoted values). However, the similarity of $\alpha, \beta, T_U,$ and $Q_o$ in materials whose microscopic structure ranges between impurities in ordered crystals to completely disordered amorphous glasses, is really striking, and suggestive of a fundamental mechanism dictating the low temperature characteristics of disordered solids. Note that above $T_U$ things change rapidly - one finds a hump in $C_v(T)$, a plateau in $\kappa(T)$, and $Q(T)$ falls very dramatically to $Q/2\pi = l/\lambda \sim O(1)$ (see, eg., ref.5).

In this Letter we present a novel approach to the problem, relating the universal properties to the symmetry of tunneling states under local inversion. Tunneling states symmetric under inversion have intrinsically weak interaction with the phonon field, defining the relevant universal small parameter as the ratio between the elastic and Coulomb energies in the solid. Universality then arises from the generality of the symmetry argument, the robustness of the energy scales involved, and a generic gapping of the asymmetric tunneling states resulting from inter-defect correlations. Being a local mechanism, our theory may shed light also on the question of the microscopic structure of amorphous solids.

Theoretical analyses of the universality began with the influential "Standard Tunneling" ("ST") model6,7, in which the low-energy modes are described by a set of non-interacting localised two-level systems (TLSs), which however interact weakly with phonons. One assumes a flat broad distribution of bias energies, with a probability distribution $P(\epsilon, \Delta_o) \sim P_o/\Delta_o$ over tunneling amplitude $\Delta_o$ and bias energy $\epsilon$. The central dimensionless parameter of the ST model is the "tunneling strength" $C_o = P_o(\gamma^2/\rho c^2)$, where $\gamma$ is the defect-phonon coupling, $\rho$ the mass density, and $c$ the phonon velocity; this model gives $C_v(T) = \alpha_o T$, $\kappa(T) = \beta_o T^2$ with $\beta_o \propto 1/C_o$, and $Q_o = 2/\pi C_o$, so that $\lambda/l = \pi^2 C_o$; and $l(T), \lambda(T)$ are both $\propto 1/T$. Thus $C_o$ dictates phonon attenuation, internal friction, and thermal conductivity.

Experimentally, $C_o \approx 10^{-3}$ in most materials, even though $\rho c^2$ varies by four orders of magnitude. There is no reason for this remarkable result to hold in the ST model, and for this reason most later theories4,8,9,10,11,12,13, recognising the importance of inter-defect
interactions, have tried to explain universality as a consequence of these. However this leads to a further problem, because in such theories the density of states \( n(E) = \rho c^2/\gamma^2 = P_o/C_o \). Typically \( P_o \sim 0.1n(E)^{2.14.15} \). This then implies \( C_o \sim 0.1 \), a huge discrepancy with experiment. Such problems have led some to argue\(^4.10.11\) that the defect modes may not be TLSs at all, but something quite different.

![Structure of crystals doped with impurities. (a) The 3-dimensional structure of the alkali halide KBr with two CN impurities, which align along one of the 3 crystal axes (distorting the nearby lattice). (b) A 2-dimensional system in which a small impurity can occupy one of the 4 positions shown by red and yellow circles; states of the same colour are related by inversion symmetry. The low-energy states of this 4-state system are isomorphic to those of an orientational impurity like CN which can rotate 90° between a "red orientation" and a yellow one, and flip through 180°, whilst keeping the same colour. States of the same colour form a "\( \tau \)-pair", and transitions between the two states of the \( j \)-th \( \tau \)-pair are generated by the operator \( \hat{\tau}_x^j \). Transitions between states having two different colours are mediated by the \( S \)-spin operators.](image)

Almost all these approaches have assumed glassy behaviour and/or interactions must be associated with universality. However amorphous systems without universality have been found\(^16\), and experiments in ion-implanted crystalline Si showed that universality was unrelated to the degree of amorphicity\(^17\). Universal properties have also been observed in the ferroelectric phase\(^18.19\), and recent evidence suggests that strong random strain fields acting on tunneling defects, rather than interactions between the defects, are responsible for universality\(^15.20.21\). This is most obvious for impurities in otherwise crystalline hosts. Thus,
FIG. 2: Interaction of $S$- and $\tau$-spins with elastic strain. In (a) we show the lattice distorted by a passing phonon; this breaks the degeneracy between states of different colours (i.e., it couples to the $S$-spins), but not between states of the same color (i.e., it does not couple directly to the $\tau$-spins, to first order in the gradient of the lattice displacement). In (b) and (c) we see how the degeneracy between 2 states of a $\tau$-pair is broken either in a system in which there is no long-range lattice structure (i.e., an amorphous system) or by the presence of a nearby impurity in a lattice. In this case the $\tau$-pair does interact, albeit weakly, with a strain field (shown here as a passing phonon).

In pure $KBr$ doped with $CN$ impurities, universality requires very high $CN$ concentration $x$ ($x > 0.2$); the impurities then create sufficiently strong random strain fields. However $CN$ impurities in $(KBr)_y(KCl)_{1-y}$ (where strong random strains already exist) give universality for very small $x$; but now $Q_o$ is $x$ dependent, and for $x \ll 1$ one finds $Q_o \gg 10^3$. Thus one can argue\textsuperscript{22} that experiments on universality should focus on disordered crystals or crystals with impurities, rather than on amorphous systems.
FIG. 3: Densities of states of $\tau$ and $S$ TLSs. General behaviour of the mean field densities of states $n^o_S(E)$, $n^o_\tau(E)$, and the true densities of states $n_S(E)$, $n_\tau(E)$, once inter-spin correlations are included. These results are shown as functions of energy (measured in temperature units) for a typical value of $J_o \sim 300$ K; both axes are plotted logarithmically. The sharp gap-like feature in $n_S(E)$ appears at a temperature $T_U \sim gJ_o$, where the $\tau$-spin levels first appear. $T_U$ is to be identified with the crossover temperature to universality. $n_\tau(E) \approx n^o_\tau(E)$ is nearly unchanged by inter-spin correlations, see text.

We now outline our theory for disordered crystals, and without loss of generality we consider the well studied example of $KBr : CN$. The applicability of our theory to amorphous solids is discussed at the end of the Letter. Fig. 1 shows $KBr : CN$, with the $CN$ impurities oriented along the 3 $KBr$ crystal axes. Each $CN$ then has 6 degenerate states, which divide into 3 "$\tau$ pairs", of states related to each other by inversion. Transitions (90° rotations) between different pairs are mediated by a pseudospin $S$. For simplicity we consider $\tau$ and $S$ to be spin-half Pauli operators (see Supplementary Information).

$S$-spins differ crucially from $\tau$-spins in their interaction with an elastic strain field $u_{\alpha\beta}(r)$. An isolated $S_j$ interacts directly with this field, whereas in a pure lattice an isolated $\tau_j$ can interact only with the gradient of the strain, inversion symmetry rules out any direct interaction (see Fig. 2). However disorder breaks the exact inversion symmetry of the $\tau$-spins, inducing a weak direct interaction with the strain field. The TLS-phonon interaction
FIG. 4: The true $S$-spin density of states $n_S(E)$, as a function of energy for different short-distance behaviours of the interaction. We normalise $n_S(E)$, defining $P(E) = n_S(E)/n_S(\bar{E})$, where $\bar{E}$ is an energy a few times larger than $T_U$; essentially $n_S(\bar{E})$ is the $S$-spin density of states obtained if one includes $S - S$ correlations but ignores $S - \tau$ correlations, so $P(E)$ measures the effect of $S - \tau$ correlations. We use the interaction form $J_{ik}^{S\tau} = c_{ik}gJ_o/(R_{ik}^3 + a_o^3)$ discussed in the text, where $c_{ik}$ is a random variable and $a_o$ is a short distance cutoff parameter. Then curves for $P(E)$ are shown for different $a_o$ (in units of $\bar{a}$), where $E$ is measured in units of $E_\tau$, the calculated average energy for $\tau$ excitations. $T_U$ is defined by the energy at which $n_\tau(E)\bar{\gamma}_w^2 = n_S(E)\gamma_s^2$, i.e. by $P(E) \approx 5g$. Impurity concentration is $x = 0.5$ and lattice size is $13^3$ cells (giving $\sim 4400$ TLSs). Similar results are obtained for $x = 0.2$ (see Supplementary Information). For both concentrations we find that $T_U \approx 0.2E_\tau$ for $1 < a_o < 6$, i.e. $T_U \sim gJ_o$.

then takes the form (see Supplementary Information)

$$V_{d\phi} = \sum_j \sum_{\alpha,\beta} [\gamma_s^{\alpha\beta} S_j^z + \gamma_w^{\alpha\beta} \tau_j^z + \eta \delta^{\alpha\beta}] u_{\alpha\beta}(r_j) \quad (1)$$

Here $\gamma_s \approx E_C$, where $E_C \approx 5$eV is the typical Coulomb energy in a solid; and the "volume energy distortion" parameter $\eta$ is comparable to, and usually somewhat smaller than $\gamma_s$. The parameter $\gamma_w(r_j)$ varies from site to site, and depends on strain disorder. We define a parameter $\bar{\gamma}_w$, the mean variance of $\gamma_w(r_j)$; it increases with disorder from zero for a crystalline host, saturating for a strongly-disordered strain field to $\bar{\gamma}_w \approx E_\Phi$, where $E_\Phi \sim 0.1$eV is the typical elastic energy in a solid. Our estimates for $\gamma_s$ and $\gamma_w$ are further discussed in the Supplementary Information, and are in agreement with recent ab-initio and
DFT calculations on KBr:CN\textsuperscript{25}. The small dimensionless parameter \( g = \gamma_{\text{w}} / \gamma_{\text{s}} = E_{\phi} / E_{\text{C}} \sim (1 - 3) \times 10^{-2} \) measures the deviation from local inversion symmetry caused by disorder, and plays a crucial role in what follows. Note that \( \tau \)-spins scatter phonons very weakly compared to \( S \)-spins, down by a factor \( g^2 \sim 10^{-4} - 10^{-3} \).

The defect-phonon interactions immediately generate phonon-mediated interactions between all the spins\textsuperscript{24,26}, along with random static strain fields acting on them\textsuperscript{24}. The \( S - \tau \) system then has an effective interaction Hamiltonian of form

\[
H_{\text{S\tau}} = \sum_{ij} [J_{ij}^{\text{SS}} S_i^z S_j^z + J_{ij}^{\text{S\tau}} S_i^z \tau_j^z + J_{ij}^{\text{\tau\tau}} \tau_i^z \tau_j^z] \quad (2)
\]

plus a random field term \( H_{\text{RF}} = \sum_j [h_j^S S_j^z + h_j^\tau \tau_j^z] \). We now show how the above Hamiltonian necessarily gives the smallness and universality of the tunneling strength \( C_0 \).

The interaction terms \( J_{ij}^{ab} \), with \( a, b = S \) or \( \tau \), can be written in the form \( J_{ij}^{ab} \approx J_o^{ab} c_{ij}^{ab} f(R_{ij} / \bar{a}) \), where the \( c_{ij}^{ab} \sim O(1) \) are complicated functions of angle\textsuperscript{24,26}, and the radial functions \( f(R_{ij}) \) depend on distance \( R_{ij} \) like \( (\bar{a}/R_{ij})^3 \) when \( R_{ij} \gg \bar{a} \), \( (\bar{a} \text{ being the lattice constant}) \). The three characteristic interaction energy scales \( J_o^{\text{SS}} \), \( J_o^{\text{S\tau}} \), and \( J_o^{\text{\tau\tau}} \) are related to each other by \( J_o^{\text{\tau\tau}} \approx g J_o^{\text{S\tau}} \approx g^2 J_o^{\text{SS}} \), implying a single fundamental interaction scale \( J_o \equiv J_o^{\text{SS}} \). Now \( J_o = \gamma_s^2 / \rho c^2 R_0^3 \sim 300 \text{ K} \), (where \( R_0 \) is the typical distance between nearby TLSs), and it does not vary dramatically between different solids. This experimental fact\textsuperscript{2,14} is also part of the universality picture. The random field strengths are also governed by \( J_o \) and \( g \); their typical strengths are \( h_j^S \sim J_o \) and \( h_j^\tau \sim g J_o \)\textsuperscript{24}.

In a naive mean-field analysis the effect of the interactions in equation (2) and the random fields is to spread the \( S \)-spin levels and \( \tau \)-spin levels over energy ranges \( J_o \) and \( g J_o \), respectively. One then gets mean field densities of states \( n_S^\phi(E) \), \( n_\tau^\phi(E) \) of Gaussian form, with \( n_S^\phi \sim 1/J_o R_0^3 \) for \( E < J_o \), and \( n_\tau^\phi \sim 1/g J_o R_0^3 \) for \( E < g J_o \) (see Fig. 3). Note that even though \( n_\tau^\phi(E) \gg n_S^\phi(E) \) for \( E < g J_o \), by a factor \( 1/g \), the \( S \)-spins still dominate the phonon scattering, because their scattering rate is far higher (by a factor \( 1/g^2 \)). Thus in mean field theory, the \( S \)-spins dominate the acoustic properties at all energies.

However this mean field picture is radically altered by inter-defect correlations. We summarise the changes as follows (see Fig. 3): (i) the true \( S \)-spin density of states \( n_S(E) \) now shows a slow fall-off from the mean-field result for \( g J_o < E < J_o \); then, for \( E < g J_o \sim 3 - 10 \text{ K} \), it falls precipitously, so that for energies \( \ll g J_o \), the \( S \)-spin states have essentially disappeared. (ii) The true \( \tau \)-spin density of states \( n_\tau(E) \) is hardly altered - it does show
a weak dip, but only below the much lower energy $J_o^{\tau\tau} \sim g^2 J_o \sim 0.1 K$. (iii) Finally, the phonon spectrum is only weakly altered.

The energy $g J_o$ now acquires a physical meaning - it is the energy below which the $\tau$-spin levels appear, and the $S$-spin levels disappear. This rapid crossover is responsible for the abrupt change in the system properties at a temperature $\sim g J_o$, which we will now identify with $T_U$. The mechanism governing the catastrophic reduction in $n_S(E)$ below $T_U$ is a more elaborate version of the "Coulomb gap" mechanism which exists in disordered conductors (adapted here to the case of 2 different spins and to dipolar interactions). Level repulsion caused by the inter-spin correlations forces states away from low energy - the strength of this effect depends both on the strength of the interactions, and the density of states of the levels. The effect on $n_\tau(E)$ is extremely small, because there are very few low-$E$ $S$-states, and because the $\tau - \tau$ interactions are so weak (see Supplementary Information for a rigorous derivation). The $S - S$ interactions are stronger and are responsible for the slow fall in $n_S(E)$, but the $\tau$-spin levels have a huge effect on the rather small number of low-energy $S$-levels because their density is so high.

To evaluate $n_S(E)$ we assume a form $J^{S\tau}_{ik} = g c_{ik} J_o / (R_{ik}^3 + a_o^3)$, which accounts for the elastic interactions at short distances (other short-distance forms produce similar results). We define $T_U$ by the condition $\bar{\gamma}_w^2 n_\tau(T_U) = \gamma_s^2 n_S(T_U)$, dictating the crossover between $\tau$ dominated and $S$ dominated phonon attenuation. The key result of this calculation (see Fig. 4 and Supplementary Information) is that although the shape of $n(S)$ depends on $a_o$, the universal regime exists for any value of the length scale $a_o$, and $T_U$ is only weakly dependent on it for typical values $a_o/\bar{a} \sim 1 - 6$; it is $\approx 0.2 \langle E_\tau \rangle \sim g J_o$, where $\langle E_\tau \rangle$ is the calculated average excitation energy of the $\tau$ TLSs.

Thus, below $T_U$ one only sees a set of $\tau$-spins, weakly coupled to each other and to phonons. Above $T_U$ the $S$-spins dominate, along with a plentiful supply of phonons, to which they couple strongly. Only in the crossover regime around $T_U$ do $S - \tau$ interactions play any role; and although their detailed form influences the shape of this crossover, it does not affect the physics above and below $T_U$.

Below $T_U$ the system of $\tau$-spins behaves essentially as a set of non-interacting tunneling states, which by symmetry are intrinsically TLSs. Thus we may now analyse this system using the parameters defined in the ST model. Having $n_\tau(E) = \rho c^2 / \bar{\gamma}_w \gamma_s$ and $P_o \approx 0.1 n(E)$ we obtain $C_o^{\tau} \approx 0.1 n_\tau(E) \bar{\gamma}_w^2 / \rho c^2 \approx 0.1 \bar{\gamma}_w / \gamma_s \approx 0.1 g \approx 10^{-3}$. In comparison to $S$ (asymmet-
ric, or standard) TLSs, phonon scattering by each \( \tau \) TLS is smaller by a factor of \( g^2 \), and their DOS is larger by a factor of \( 1/g \), resulting in \( C^\tau_o \approx gC^S_o \). Thus we recover universality, now with the correct experimental value for \( C_o \).

The universality then occurs because neither the parameter \( g = E_\Phi/E_C \) nor the energy scale \( J_o \) vary much between different strongly-disordered materials. Both the crossover energy \( T_U \sim gJ_o \) and the parameter \( C^\tau_o \) determining the universal properties must then be very similar for most systems; their small size comes from the small value of \( g \). We stress that glassiness, and the mechanisms leading to it, are clearly not required in our theory. If glassiness does occur, it has no relation to universality here except insofar as the glass temperature \( T_G \sim J_o \) (so that \( T_U \sim gT_G \)).

Finally, let us go beyond the model of a lattice system distorted by impurities, including some features omitted above.

(i) **Mixed Crystals:** Here the \( \tau \)-impurity concentration \( x \ll x_c \), but the host is now strongly disordered (e.g., in KBr:KCl with CN impurities). Thus strain disorder is strong even when \( x = 0 \); it comes now from the random field exerted by the random host atoms, operating through the volume term in equation (1), rather than from interactions with \( S \) impurities. The model then predicts a reduction in \( n_\tau \) by a factor \( x/x_c \), so that \( C_o \approx 0.1gx/x_c \), in agreement with experiment. Conversely, in dilute single crystals \( \gamma_w \) is typically small (\( \propto x^{4/3} \), see Supplementary Information), has a broad distribution, and is correlated with the TLS energy: then universality is lost.

(ii) **Electric Dipole Interactions:** Both \( \tau \) and \( S \) defects will have electric dipole moments, leading to electric dipole mediated interactions whose strength at nearest impurity distance is \( J_{ee} \). Thus \( J_o > J_{ee} \) almost always, but the ratio \( J_{ee}/J_{S\tau} = J_{ee}/gJ_o \) can be small or large, and typically \( J_{ee} > J_{\tau\tau} = g^2J_o \). If \( J_{ee} < gJ_o \), then all the arguments above go through, but now the \( \tau - \tau \) interaction strength is \( J_{ee} \), and this will change the temperature at which the very weak dipole 'depression' appears in \( n_\tau(E) \) to \( J_{ee} \). Such dipole depressions in the density of states are seen in some experiments below 1 K. On the other hand if \( J_{ee} > gJ_o \), electric dipole interactions also change the \( S - \tau \) interactions. This does not change the basic picture, but now one finds \( T_U \sim J_{ee} \), and \( C_o \) is reduced to \( C_o \sim g^2J_o/J_{ee} \).

(iii) **Amorphous systems and glasses:** While amorphous materials show no long range order, local order with a lattice bond coordination number does exist except at defect sites, and inter-site distances are hardly altered from lattice values. This is true even in strongly
amorphous Si, where $\delta a/\bar{a} \sim 0.03$ for nearest neighbours, and $\sim 0.1$ for 2nd nearest neighbours ($\bar{a}$ is the regular lattice value as before) and the nearest-neighbour bond angle differs by a maximum of 10%. Now our results depend essentially on the distinction between $S$ and $\tau$ defects, and rely on local properties of the system (e.g. contributions to $\gamma_w$ from disorder diminish like $\sim 1/r^4$; see Supplementary Information). Thus neither $J_o$ nor $g$ are strongly affected by amorphicity.

This allows us to make some clear predictions for amorphous systems. First, if inversion-symmetric TLSs do exist therein, then our model predicts amorphous systems will also show universality with similar values of $T_U$ and $C_o$. Second, one should find two types of defect excitations, differing by $\sim 10^2$ in their coupling to phonons, and with the densities of states given in Figs. 3, 4. Conversely, universality should be absent in amorphous systems without inversion-symmetric TLSs.

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Additional information

Supplementary information accompanies this paper at www.nature.com/naturephysics.

I. SUPPLEMENTARY INFORMATION

A. Mapping to the system of $S$ and $\tau$-spins

We consider first the 2-dimensional example given in the text, where the orientable $CN$ impurity can lie in one of 4 directions in the host lattice. A similar problem is posed by a simple point impurity which may take up one of 4 off-centre positions in the host. One may now choose 2 pairs of states from these 4 states; members of each pair are related by inversion symmetry, and we refer to these as $\tau$-spins; they are described by the Standard Tunneling (ST) model. For a single site in the 2-dimensional host, there are two $\tau$-spins, $\tau_\nu$, with Hilbert spaces $\Upsilon_\nu$, $\nu = 1, 2$. Transitions between the 2 states of a given $\tau$ spin (corresponding to $180^\circ$ flips for the $CN$ impurity, or tunneling between 2 positions on opposite sides of the centre of symmetry, for simple impurities) are mediated by $\hat{\tau}_x$. There are also transitions which take the system between $\Upsilon_1$ and $\Upsilon_2$ (these correspond to $90^\circ$ rotations for the $CN$ impurities, and hopping between nearest-neighbour off-centre points for a simple impurity). Now if we ask what is the interaction between the defect and a phonon field, we find that it must have the form
\begin{equation}
\Gamma_n^{\alpha\beta}(r_j) u_j^{\alpha\beta} + \zeta_{\alpha\beta\gamma}(r_j) \partial_\gamma u_j^{\alpha\beta} \tau_j^z
\end{equation}

where \( n = 1 \ldots l \) enumerates the pairs of states (for a 3-d cubic alkali halide lattice, there are \( l = 3 \tau \) spins per site). As noted in the text, the strain field couples directly to transitions between the subspaces, but only its gradient can couple to individual \( \tau \) variables, because of the inversion symmetry. For \( n = 2 \) we can write

\begin{equation}
\Gamma_n^{\alpha\beta}(r_j) - \Gamma_n^{\alpha\beta}(r_j) = \gamma_{j}^{\alpha\beta} [\hat{S}_j^z - \hat{S}_j^z].
\end{equation}

The resulting analogy to a system of 2 spin half particles is not exact. For example, the operator \( \hat{S}_j^x \hat{\tau}_j^x \) would flip the spin through 180° and then rotate it through 90° in one direction, but this is then equivalent to the rotation by 90° in the opposite direction. However, in this paper we are only interested in evaluating the effective interactions between defect degrees of freedom, for which we only need the Ising variables \( \hat{S}_j^z, \hat{\tau}_j^z \). Also, the reduction of the larger \( n \) space of the S degree of freedom to a spin half TLS does not affect the physics below \( T_U \), and introduces only small quantitative change to \( T_U \) itself, because of the sharp energy dependence of \( N_S(E) \) at the relevant low energies (see Fig. 4 in the main text).

**B. TLS-phonon interactions and the random strain field**

We wish to show that in the presence of disorder, \( \tau \)-TLSs acquire a finite effective interaction with the first derivative of the phonon displacement, given by

\begin{equation}
\sum_j \sum_{\alpha,\beta} \gamma_{\alpha\beta}(r_j) u_{\alpha\beta}(r_j) \tau_j^z
\end{equation}

where \( u_{\alpha\beta}(r_j) = (\partial X_{j\alpha}/\partial x_{j\beta}) \) is the strain tensor, defined in the usual way as the gradient at site \( j \) of the lattice position vector \( X_j \). To derive this we consider the interaction Hamiltonian between phonons and defects, of which the relevant terms are

\begin{equation}
H_{\text{int}} = \sum_j \left[ \sum_{\alpha} \eta(\mathbf{r}_j) u_j^{\alpha\alpha} + \sum_{\alpha,\beta} \gamma_{\alpha\beta}(\mathbf{r}_j) u_j^{\alpha\beta} S_j^z + \sum_{\alpha,\beta,\gamma} \zeta_{\alpha\beta\gamma}(\mathbf{r}_j) \partial_\gamma u_j^{\alpha\beta} \tau_j^z + \sum_{\alpha,\beta,\gamma,\delta} C_{\alpha\beta\gamma\delta}(\mathbf{r}_j) u_j^{\alpha\beta} u_j^{\gamma\delta} \right].
\end{equation}

The first term is a volume distortion term. The second term denotes the interaction of the \( S \) degree of freedom with the phonon strain, and is just that discussed above (see Eq. (S2)); both terms have been discussed exhaustively in the literature for crystals (see e.g. Refs. [1,2]).
FIG. S1: Interaction of the symmetric TLSs with the gradient of the strain. The degeneracy of states related by inversion symmetry is broken by the second derivative of the phonon displacement.

The third term denotes the interaction of the $\tau$-spins with the gradient of the phonon strain (see Fig. S1); it was derived for a crystal in ref. 2. The last term, 2nd-order in the strain, includes the modification of the elastic modulus by the impurities, and is given in its general form. In this microscopic Hamiltonian there is no direct interaction between the $\tau$-spins and $u_{j}^{\alpha\beta}$; as noted in the text and in ref. 2, this is forbidden by symmetry in a pure crystal.

A simple way to estimate the effect of strain disorder on the interaction between $S$ and $\tau$ impurities, is to re-derive this interaction with a 3rd impurity introduced into the calculation. Without this 3rd impurity, the effective $\tau_{i}^{z}S_{j}^{z}$ between two distant impurities is $\sim 1/R^{4}$, in 2nd-order perturbation theory\textsuperscript{2}. We now calculate the effective $\tau_{i}^{z}S_{j}^{z}$ interaction in third order in $H^{\text{int}}$, and deduce $\gamma_{w}$ from the coefficient of the term behaving as $1/R^{3}$ for large distances\textsuperscript{3}. We assume the 3rd impurity $k$ is much closer to impurity $i$, to model deviation from inversion symmetry resulting from strong disorder strain.

We write the Fourier transform of the lattice displacement field as

\begin{equation}
X_{i}(x) = \frac{1}{\sqrt{N}} \sum_{q,\mu} X_{q,\mu} e_{q,\mu,\alpha} e^{i q x}
\end{equation}

where $e_{q,\mu,\alpha}$ is a phonon polarization index. We then minimise the total potential energy, i.e. the sum of the bare phonon potential energy plus the interaction term above, to find
the resulting distortion in the lattice. This distortion is then found to be

$$\delta X(x) = \frac{1}{2} \sum_{q \mu} (\delta X_{q \mu} e^{iqx} + H.c.), \quad (S6)$$

where we have

$$\delta X_{q,\mu} = \frac{1}{\sqrt{NM \omega_{q\mu}^2}} \left( \sum_{\gamma,\delta,\eta} \zeta_i^{\gamma \delta \eta} e_{q \gamma} q \delta q \eta e^{-iqx_i^\gamma} + i \sum_{\alpha \beta} \gamma_j^{\alpha \beta} e_{q \mu a} q \beta e^{-iqx_j^\gamma} S_j^z \right). \quad (S7)$$

The effect of this distortion on the 3rd impurity comes only from the fourth term in $H_{int}$, which can be written as

$$\sum_{\rho \phi \chi \psi} C_{\rho \phi \chi \psi}(r_k) \frac{\partial \delta X_{k \rho}}{\partial x_k} \frac{\partial \delta X_{k \chi}}{\partial x_k} \quad (S8)$$

where

$$\frac{\partial \delta X_{k \rho}}{\partial x_k} = \sum_{q \mu} \frac{1}{NM \omega_{q \mu}^2} \left( \sum_{\gamma,\delta,\eta} \zeta_i^{\gamma \delta \eta} e_{q \gamma} q \delta q \eta \sin q(x_k - x_i)\tau_i^\gamma + \sum_{\alpha \beta} \gamma_j^{\alpha \beta} e_{q \mu a} q \beta \cos q(x_k - x_j)S_j^z \right) e_{q \mu \phi} q \phi. \quad (S9)$$

We now evaluate the term in Eq. (S8) proportional to $\tau_i^\gamma S_j^z$. Defining

$$A_{q \mu} = \sum_{\gamma,\delta,\eta} \zeta_i^{\gamma \delta \eta} e_{q \gamma} q \delta q \eta \tau_i^\gamma \quad (S10)$$

$$D_{q \mu} = \sum_{\alpha \beta} \gamma_j^{\alpha \beta} e_{q \mu a} q \beta S_j^z \quad (S11)$$

we find the change in the $\tau_i^\gamma S_j^z$ interaction caused by the impurity at site $k$ to be

$$E_k(i, j) = 2 \sum_{\rho \phi \chi \psi} C_{\rho \phi \chi \psi}(r_k) \sum_{q \mu} \frac{e_{q \mu \phi} q \phi}{NM \omega_{q \mu}^2} A_{q \mu} \sin [q(x_k - x_i)] \sum_{q' \mu'} \frac{e_{q' \mu' \phi} q' \phi}{NM \omega_{q' \mu'}^2} D_{q' \mu'} \cos [q'(x_k - x_j)]. \quad (S12)$$

In the acoustic approximation one finds

$$E_k(i, j) = \frac{c_{ij} \zeta_i^\gamma \gamma_j S_j^z C_k}{\rho^2 A R_{ik}^4 R_{jk}^3}. \quad (S13)$$

where $c_{ij} \sim 1$ is a complicated function of angle, which depends on the position of the impurities and takes either sign. It can then be treated as a random variable. Here the acoustic approximation is not valid, because $R_{ik} \approx \bar{a}$. However, we are only interested here in an order of magnitude, and using the fact that $R_{ik} \ll R_{jk}$ we find
\[ E_k(i, j) \approx \frac{\gamma_w \gamma_s}{\rho c R_{ij}^3} \]  \hspace{2cm} (S14)

where

\[ \gamma_w \approx \frac{\zeta C}{\rho c R_{13}^4}. \]  \hspace{2cm} (S15)

Here \( \zeta, C, \) and \( \gamma \) stand for typical values.

Actually, all impurities contribute to the deviation from inversion symmetry at site \( i \), and therefore to \( \gamma_w \). However, since this contribution depends on \( 1/R_{ik}^4 \), and has random sign, \( \gamma_w \) is strongly dominated by the contributions from the closest impurities. Thus we can assume that impurity \( k \) is a nearest neighbour impurity to impurity \( i \).

\[ \text{FIG. S2: The true } S\text{-spin density of states } n_s(E), \text{ as a function of energy for different short-distance behaviours of the interaction. Details are as in Figure 4 in the main text, only here } x = 0.2 \text{ and lattice size is } 18^3 \text{ cells (ie., } \sim 4650 \text{ TLSs).} \]

Let us now estimate the magnitude of \( \gamma_w \). First, \( C \cdot (\delta a/\bar{a})^2 \approx \delta M \omega^2 (\delta a)^2 \), where \( \delta a \) denotes a typical strain, and the latter expression is the difference in kinetic energy of the impurity compared to the host ion. Approximating \( \delta M \approx M \) and \( \omega \approx c/\bar{a} \) we obtain \( C \approx Mc^2 \). Since \( R_{13}^4 \approx \bar{a}^4 \bar{n}^{-4/3} \), where \( \bar{n} \) is the dimensionless impurity concentration, we get \( \gamma_w \approx \bar{n}^{4/3} \zeta /\bar{a} \).

Let us now estimate \( \zeta \). Since \( \zeta \) is the coefficient of the second derivative of the displacement, it involves interaction with next nearest neighbors (see Fig. S1). Thus, \( \zeta \cdot \delta a/\bar{a}^2 \approx E_\Phi \cdot \delta a/\bar{a} \) so that \( \zeta \approx \gamma_s \bar{a} E_\Phi /E_C \). Thus, for \( \bar{n} \approx 1 \) we find \( \gamma_w \approx \gamma_s E_\Phi /E_C \).
Note, that the ratio between the typical strain in a strongly disordered or amorphous solid to the interatomic distance, is $\delta a/\bar{a} \approx E_\Phi / E_C$, and therefore $g \equiv \gamma_w / \gamma_s \approx \delta a / \bar{a}$.

The calculation above can be similarly done for the effective random field exerted by the first term in the interaction Hamiltonian \[ S1 \], with no reference to the S-TLSs.

1. **Dilute impurities in a lattice**

The reduction of $x$ below $x_c \approx 0.2$ has two main effects. First, the typical strength of the interaction of the $\tau$ TLSs with the lattice becomes smaller, $\gamma_w \approx g \gamma_s n^{1/3}$. This, together with the dependence of the typical TLS-TLS interaction on $n$, would limit the universal regime to $T < n^{7/3} T_U$. However, even within this much smaller temperature regime one can not expect universal behavior. This is since the variance of $\gamma_w$ becomes large, and correlated with the TLS energy: strong local strains acting on the $\tau$ spins from nearby $S$-impurities increase both $\gamma_w$ and $J^{S\tau}$. Such a correlation invalidates the assumptions of the ST model, and results in a temperature dependence of the phonon attenuation. In the mixed crystals (e.g. KBr:KCl with CN impurities) the situation is different, as the random fields experienced by the $\tau$ TLSs are $x$ independent. Thus, the condition required by the ST model that the impurities have similar interaction with the lattice, and that these are not correlated with their excitation energies, are satisfied. Furthermore, the disorder in the energies $E_\tau$ is dictated by the random fields, and is therefore independent of $x$. The only effect of having small $x$ is the reduction in $n(\tau)$, and we therefore find that universal properties exist in the mixed crystals, albeit with a tunneling strength $C \approx 0.1 g x / x_c$.

C. **Dipolar gap for the interacting $S$-$\tau$ system**

In this section we calculate the gap for single particle excitations for both the S and $\tau$ TLSs. Consider the Hamiltonian

$$\sum_{ij} J^{SS}_{ij} S^z_i S^z_j + \sum_{ij} J^{S\tau}_{ij} S^z_i \tau^z_j + \sum_{ij} J^{\tau\tau}_{ij} \tau^z_i \tau^z_j \quad (S16)$$

where $J^{ab}_{ij} = c^{ab}_{ij} J^{ab}_o / (R^3_{ij} + a^3_o)$, $a, b = S\tau$, $c^{ab}_{ij}$ is a random number of order unity, $J^{\tau\tau}_o = g J^{S\tau}_o = g^2 J^{SS}_o$, and $g \ll 1$ ($g \approx 0.01 - 0.03$ in our case). We introduce here a cutoff length $a_o$ for the interaction. Rounding the diverging interaction at small distances is crucial for
the calculation of the gap of the S-TLSs, but its functional form and the exact value of the cutoff parameter are only of small quantitative relevance.

1. gap of $\tau$ impurities

Consider first the gap exerted by the S spins on the $\tau$ spins, neglecting the $\tau - \tau$ interaction (third term). Following Efros and Shklovskii,

$$n_\tau(E) = n^o_\tau(E) \prod_j \Theta(E_\tau + E_{Sj} - 2U_j) \equiv n^o_\tau(E) P(E_\tau). \quad (S17)$$

Here $n^o_\tau(E) \approx 1/gJ_o$ corresponds to the ungapped DOS, and $U_j$ is the interaction between the given $\tau$ spin and the $j’th$ S spin. We would like to show that there is no appreciable reduction of the $\tau$ DOS, at least outside a window which is exponentially small in $g$. We proceed by assuming a series of simplifying conditions, all leading to a reduction of $P(E_\tau)$ below its actual value, and obtain our final result in form of an inequality.

We first overestimate $n_S(E) = 1/J_o$ for all $E < J_o$, neglecting their reduced DOS at low energies. We then enumerate the S spins according to their distance from the $\tau$ impurity. Since there are $j$ impurities within a volume $(r_j)^3$, the maximum interaction is given by $U_j^{max} = gJ_o/j$. We assume that all interactions have this maximum value, taking the short distance cutoff $a_o = 0$ and $c_{ij}^{S\tau}_o = 1$. Under the above assumptions, and defining $E_\tau = \epsilon$, we obtain for $\epsilon \ll gJ_o$

$$P(\epsilon) = \prod_j \int_{2gJ_o/\epsilon - \epsilon}^{2gJ_o/\epsilon} d\nu \frac{1}{J_o} \prod_j \left( 1 - \frac{2gJ_o/j - \epsilon}{J_o} \right). \quad (S18)$$

Defining $\epsilon' \equiv \epsilon/2gJ_o$ we obtain

$$P(\epsilon') = \prod_j \left[ 1 - 2g \left( \frac{1}{j} - \epsilon' \right) \right] > \prod_j \left( 1 - \frac{2g}{j} \right) > \prod_j \frac{1}{1 + 4g/j}. \quad (S19)$$

Multiplying the denominators, and expanding in a series in $g$, we see that
\[
\prod_{j}^{1/\epsilon'} \left( 1 + \frac{4g}{j} \right) < 1 + \sum_{k} (4g \ln 1/\epsilon')^k.
\]  
(S20)

and therefore

\[
P(\epsilon') > 1 - 4g \ln 1/\epsilon'.
\]  
(S21)

i.e. \(P(\epsilon') \approx 1\) for \(\epsilon' > \exp(-1/8g)\).

Below we will argue that the S impurities are strongly gapped themselves, thus also the above small correction at \(g^2J_o < E_\tau < gJ_o\) is probably an overestimate.

To calculate the gap that the \(\tau\) impurities exert on themselves, one can repeat the same arguments as above, with \(J_{o}^{SS} \equiv J_{o}\) replaced by \(J_{o}^{S\tau} \equiv gJ_{o}\) and \(J_{o}^{S\tau} \equiv g^2J_{o}\). Eq.
(S21)
 is thus reproduced as an inequality. However, since unlike the S impurities, the \(\tau\) impurities are not strongly gapped, one can recover the line of arguments with \(\approx\) instead of \(>\), and conclude that

\[
P_{\tau\tau}(\epsilon'') \approx 1 - c \ln 1/\epsilon''.
\]  
(S22)

where \(\epsilon'' \equiv \epsilon/g^2J_o\) and \(c \approx g\). Thus, the correction to the DOS at \(E < g^2J_o\) is reduced by the small parameter \(g\), which is the ratio between the \(\tau - \tau\) interaction and their energy disorder, and is also the relevant small parameter in our theory. Experimentally, the dipole gap is indeed seen at energy scales comparable to the \(\tau - \tau\) interactions\(^{4}\), and its magnitude is considerably reduced.

2. gap of S impurities

The calculation of the gap of the S impurities is more subtle, since there is no small parameter, and the result is of order unity. Furthermore, the correlation between the interaction and \(\tau\) energies is crucial, and must be taken into account. We therefore solve the problem using a numerical simulation. Neglecting the third term in Eq. (S16) the values of \(E_\tau\) are dictated by the second term. We are interested in \(P(E_S)\), defined by

\[
n_S(E) = n_S(\bar{E}) \prod_j \Theta(E_S + E_{\tau_j} - 2U_j) \equiv n_S(\bar{E})P(E_S).
\]  
(S23)
Here $E$ is an energy a few times larger than $T_U$ and $U_j$ is the interaction between a given $S$-TLS and $\tau_j$. We therefore use the following algorithm:

We choose a lattice size and concentration, and randomly distribute the impurities in the lattice. Each impurity has an $S$ spin and a $\tau$ spin. For a given $a_o$, the interaction is given by $U_{ij} = c_{ij} S_i^z \tau_j^z J_{0}^{S\tau} / (R_{ij}^3 + a_o^3)$. We define $c'_{ij} \equiv c_{ij} S_i^z$, where $c'_{ij}$ is chosen from a random distribution with width unity. We then obtain $U_{ij}$ and $E_{\tau_j} \equiv \sum_i U_{ij}$ with their essential dependence, but with no reference to the spin configuration of the $S$ TLSs. We then flip the $\tau$ spins where $E_{\tau} < 0$, to have a positive excitation energy. The $U_{ij}$'s are accordingly redefined. For each $S_i$ we then calculate $E_{S_i}^{\text{max}}$, the maximal $E_{S_i}$ satisfying $\prod_j \Theta(E_{S_i} + E_{\tau_j} - 2U_{ij}) > 0$. We then obtain $P(E_S) = N(E_{S_i}^{\text{max}} < E_S) / N(S)$, the ratio between the number of spins with $E_{S_i}^{\text{max}} < E_S$ to the total number of spins. The results for $x= 0.5$ are given in Fig. 4 in the main text, and for $x= 0.2$ in Fig. S2.

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