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Two-level systems and mass deficit in quantum solids

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We study a disordered quantum solid incorporating two-level systems in which a group of atoms (or a single atom) can experience coherent tunnelling between two different positions and demonstrate that an effective mass deficit induced by the presence of such objects can manifest itself only at relatively high frequencies and should vanish in the low-frequency limit. The crossover to the regime which can be associated with the appearance of an effective mass deficit has been observed in recent torsional oscillator experiments.

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1. Introduction. After a number of torsional oscillator experiments [1 – 3] demonstrated that at low temperatures solid ^{4}He behaves itself as if it contained a superfluid component which remains at rest when an external force is applied to the sample, an idea was put forward that these properties can be explained by the presence of two-level systems (TLSs) in which a group of atoms (or a single atom) experiences classical [4] or quantum [5, 6] tunnelling between two localized positions shifted with respect to each other. To some extent this conjecture is confirmed by the dependence of the experimentally observed mass deficit on the degree of disorder in the solid [2].

However, the analysis of Ref. [5, 6] has led to a rather paradoxical conclusion that the effective mass deficit induced by TLSs should remain non-vanishing even in the adiabatic limit including the really stationary situation of the thermodynamic equilibrium, when experimentally the system behaves as if $\rho_s = 0$ [7]. The origin of this conclusion can be traced to the assumption that the bulk velocity of a solid is a classical variable commuting with the Hamiltonian of a TLS.

In the present note we propose an alternative approach which takes into account the operator nature of different variables in a more consistent way. This allows us to show that a quantum solid with incorporated TLSs can demonstrate the presence of a pronounced mass deficit only when an external force changes fast enough, but not in a stationary situation or at very low frequencies. For comparison, we also discuss the situation when the tunnelling inside the TLSs is incoherent.

2. A solid with a quantum two-level system. Consider a solid of total mass M incorporating a TLS in which a group of atoms (or a single atom) of mass m can tunnel between two localized positions shifted by vector $\mathbf{a}$ with respect to the rest of the solid. Below we use the term "solid frame" to designate the part of the solid which does not participate in the process of tunnelling and the term "system" to describe the whole system consisting of the solid frame and the TLS.

In the absence of any interaction between the TLS and other internal degrees of freedom (for example, phonons) the quantum-mechanical Hamiltonian of such a system can be written as

$$\hat{H}_0 = \frac{1}{2M}\mathbf{P}^2 - \varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1, \tag{1}$$

where $\mathbf{P} = -i\hbar\partial/\partial\mathbf{R}$ is the operator of total momentum of the system (conjugate to the center of mass position $\mathbf{R}$), the second term describes the difference in energy (given by 2ε) between the two localized states of the TLS and the third one the process of quantum tunnelling (with amplitude J) between these two states, $\hat{\sigma}_1$ and $\hat{\sigma}_3$ being the Pauli matrices.

Naturally, the velocity of the center of mass of the system,

$$\mathbf{V} \equiv \frac{d}{dt}\mathbf{R} = \frac{i}{\hbar}[H_0, \mathbf{R}] = \frac{1}{M}\mathbf{P}, \tag{2}$$

is determined just by its total momentum $\mathbf{P}$ and is insensitive to its internal life (a particular state of the TLS). In the presence of an external force $\mathbf{F}(t)$ applied to the system as a whole (which corresponds to replacing $\hat{H}_0$ by $\hat{H} = \hat{H}_0 - \mathbf{R}\mathbf{F}(t)$), the time evolution of $\langle\mathbf{V}\rangle$ is entirely determined by relation $(d/dt)\mathbf{V} = \mathbf{F}/M$.

However, in a number of experimental situations an external force (for example, of mechanical origin) is applied not to the center of mass of the system, but to the solid frame, and one is interested in the relation between this force $\mathbf{f}(t)$ and the velocity of the solid frame[2)], $\mathbf{v}(t)$,

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[2)]Andreev calls the same variable "the solid bulk velocity" [5, 6].

which is not obliged to have exactly the same form as the relation between $\mathbf{V}$ and $\mathbf{F}$. The coordinate describing the position of the solid frame, $\mathbf{r}$, can be introduced by rewriting the definition of the center of mass position $\mathbf{R}$ as

$$M\mathbf{R} = (M - m)\mathbf{r} + m\mathbf{x} = M\mathbf{r} + m(\mathbf{x} - \mathbf{r}), \qquad (3)$$

where $\mathbf{x}$ is the position of the center of mass of the atoms (atom) forming the TLS. After replacing $\mathbf{u} \equiv \mathbf{x} - \mathbf{r}$, the displacement of the TLS with respect to the solid frame, by $-\frac{1}{2}\mathbf{a}\hat{\sigma}_3$, Eq. (3) can be rewritten as

$$\mathbf{r} = \mathbf{R} + \frac{m}{2M}\mathbf{a}\hat{\sigma}_3. \qquad (4)$$

The operator of the solid frame velocity $\mathbf{v} \equiv (d/dt)\mathbf{r}$ is then given by

$$\mathbf{v} = \frac{i}{\hbar}\left[\hat{H}_0, \mathbf{r}\right] = \frac{1}{M}\left(\mathbf{P} + \frac{mJ}{\hbar}\mathbf{a}\hat{\sigma}_2\right). \qquad (5)$$

Naturally, this equation can be also rewritten as

$$\mathbf{P} = M\mathbf{v} - \frac{mJ\mathbf{a}}{\hbar}\,\hat{\sigma}_2, \qquad (6)$$

which corresponds to splitting the total momentum of the system $\mathbf{P}$ into the two terms related respectively with the solid frame and the TLS. However it is important that in contrast to the center of mass velocity $\mathbf{V} \equiv \mathbf{P}/M$, which in the absence of external force commutes with the Hamiltonian, the velocity of the solid frame $\mathbf{v}$ is an operator which does not commute with $\hat{H}_0$.

In the presence of an external force $\mathbf{f}(t)$ applied to the solid frame the Hamiltonian of the system acquires form

$$\hat{H} = \hat{H}_0 - \mathbf{r}\mathbf{f}(t) = -\mathbf{R}\mathbf{f}(t) - h_a(t)\hat{\sigma}_\alpha \qquad (7)$$

where

$$h_\alpha(t) = \left[-J,\ 0,\ \varepsilon + \frac{m}{2M}\mathbf{a}\mathbf{f}(t)\right] \qquad (8)$$

plays the role of the effective magnetic field acting on the spin $1/2$ which can be associated with the TLS and subscript $\alpha = 1, 2, 3$ denotes the components of $\mathbf{h}$.

It follows from Eq. (5) that

$$\frac{d}{dt}\langle\mathbf{v}\rangle = \frac{\mathbf{f}}{M} + \frac{m}{M}\frac{J\mathbf{a}}{\hbar}\frac{d}{dt}\langle\sigma_2\rangle, \qquad (9)$$

which suggests that the coefficient of proportionality between $(d/dt)\langle\mathbf{v}\rangle$ and $\mathbf{f}$ can be different from $1/M$. To find a more explicit form of a TLS-induced correction to the solid frame's equation of motion one needs to express the time derivative of $\langle\sigma_2\rangle$ in terms of $\mathbf{f}(t)$.

3. Adiabatic regime. If external force $\mathbf{f}(t)$ does not depend on time or changes very slowly one needs to take into account the relaxation processes which force vector $\langle\sigma_\alpha\rangle$ to remain always parallel to h_α, from where $\langle\sigma_2\rangle = 0$. Therefore, when $\mathbf{f}(t)$ changes sufficiently slowly the processes inside the TLS make no corrections to $\mathbf{P} = M\mathbf{v}$. Thus, in the adiabatic regime the presence of the TLS cannot lead to the appearance of any difference between the real mass of the system and its effective mass observed in experiments involving the application of mechanical force.

The finite expression for the mass deficit in the stationary regime derived by Andreev [5] in the easiest way can be reproduced by calculating the average of the TLS contribution to the total momentum $\mathbf{P}$, that is of the second term in Eq. (6), with the help of the Hamiltonian,

$$\hat{H} = \frac{M}{2}\mathbf{v}^2 - \frac{mJ}{\hbar}(\mathbf{a}\mathbf{v})\,\hat{\sigma}_2 - \varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1, \qquad (10)$$

which in Ref. [5] is obtained by applying to $H_{\text{TLS}} = -\varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1$ the Galilean transformation from the reference frame in which the solid frame is at rest to the reference frame moving with velocity $\mathbf{v}$. Naturally, the application of this procedure implies that solid frame velocity $\mathbf{v}$ can be treated as a classical variable.

One can easily check that Eq. (10) differs from Eq. (1) with $\mathbf{P}$ replaced by $M\mathbf{v} - (mJ\mathbf{a}/\hbar)\hat{\sigma}_2$ only by a trivial constant term which additionally tends to zero in the thermodynamic limit, $m/M \to 0$. Thus the basic difference between the two approaches consists only in choosing whether $\mathbf{P}$ or $\mathbf{v}$ is a classical variable commuting with the Hamiltonian. The first option (adopted here) leads to $\langle\hat{\sigma}_2\rangle = 0$ and $\langle\mathbf{P}\rangle = M\mathbf{v}$, whereas the second one produces for the average of the TLS contribution to the total momentum the expression [5]

$$\langle\mathbf{P}_{\text{TLS}}\rangle = -\left(\frac{mJ}{\hbar}\right)^2 \frac{\tanh[E(\mathbf{v})/T]}{E(\mathbf{v})}\mathbf{a}(\mathbf{a}\mathbf{v}) \qquad (11)$$

with

$$E(\mathbf{v}) \equiv \sqrt{\varepsilon^2 + J^2 + (mJ/\hbar)^2\,(\mathbf{a}\mathbf{v})^2}, \qquad (12)$$

which does not respect the Galilean invariance being nonlinear in $\mathbf{v}$.

4. Finite-frequency response. When interaction of the TLS with other degrees of freedom (heat bath) is weak, the basic form of the linear response of the TLS to the application of the external force with a finite frequency, $\mathbf{f}(t) \propto \cos(\omega t)$, can be found by constructing the periodic solution of the equations describing the free evolution of the TLS [6],

$$\frac{d}{dt}\hat{\sigma}_\alpha = \frac{i}{\hbar}[H,\hat{\sigma}_\alpha] = -\frac{2}{\hbar}\epsilon_{\alpha\beta\gamma}h_\beta\hat{\sigma}_\gamma. \tag{13}$$

After diagonalizing $H_{\rm TLS} = -\varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1$ and constructing the corresponding finite-temperature density matrix $\hat{\rho} = \exp\left(-\hat{H}_{\rm TLS}/T\right)$ one obtains that in the absence of an external force

$$\langle\hat{\sigma}_\alpha\rangle^{(0)} = \left(-\frac{J}{E},\ 0,\ \frac{\varepsilon}{E}\right)\tanh\frac{E}{T}. \tag{14}$$

with $E \equiv \sqrt{\varepsilon^2 + J^2}$.

Solution of the equations for $\langle\hat{\sigma}_\alpha\rangle$ obtained by the linearization in the vicinity of $\langle\hat{\sigma}_\alpha\rangle = \langle\hat{\sigma}_\alpha\rangle^{(0)}$ gives

$$\frac{d}{dt}\langle\hat{\sigma}_2\rangle = -\frac{m}{M}\frac{\omega^2}{\omega^2-\Omega^2}\frac{\mathbf{af}(t)}{\hbar}\langle\hat{\sigma}_1\rangle^{(0)}, \tag{15}$$

with $\Omega = 2E/\hbar$. Substitution of Eq. (14) into Eq. (15) and then into Eq. (9) transforms the latter into

$$\frac{d}{dt}\langle v_i\rangle = \left[\frac{\delta_{ij}}{M} + \frac{\omega^2}{\omega^2-\Omega^2}\frac{a_i a_j}{M^2}\lambda\right] f_j(t), \tag{16}$$

where subscripts i and j denote the components of vectors in the real space and

$$\lambda \equiv \lambda(T) = \frac{m^2 J^2}{\hbar^2 E}\tanh\frac{E}{T}. \tag{17}$$

When one additionally takes into account the processes of transverse relaxation induced by the interaction with the heat bath[3)], the singularity at $\omega = \Omega$ is smeared out and the TLS-induced correction to $(d/dt)\langle v_i\rangle$ acquires also a dissipative contribution, proportional not to $\cos(\omega t)$ but to $\sin(\omega t)$. If for simplicity one assumes that the relaxation can be characterized by the same relaxation time τ (with $\Omega\tau \gg 1$) for both directions perpendicular to $\langle\hat{\sigma}_\alpha\rangle^{(0)}$, the form of the result corresponds to the replacement of $\omega^2 - \Omega^2$ in the denominator in Eq. (16) by $(\omega + i/\tau)^2 - \Omega^2$. The dissipative contribution is the dominant one when $\tau|\omega - \Omega| \ll 1$.

When the system contains many TLSs with random orientations, Eq. (16) can be replaced by $(d/dt)\langle\mathbf{v}\rangle = \mathbf{f}(t)/M_{\rm eff}(\omega)$ with

$$\frac{1}{M_{\rm eff}(\omega)} = \frac{1}{M} + \frac{1}{3M^2}\sum_n \frac{\omega^2}{(\omega + i/\tau_n)^2 - \Omega_n^2}\gamma_n a_n^2, \tag{18}$$

[3)]The interaction with the heat bath can also lead to the renormalization of J [8].

where subscript n numbers different two-level systems and we have assumed that vectors $\mathbf{a}$ have an isotropic distribution. As usual with such a notation, the real and imaginary parts of $1/M_{\rm eff}(\omega)$ describe the amplitudes of terms in $(d/dt)\langle\mathbf{v}\rangle$ which are proportional respectively to $\cos(\omega t)$ and $\sin(\omega t)$ when an external force applied to the solid frame is proportional to $\cos(\omega t)$.

It follows from the structure of Eq. (18) that when ω is much larger than all Ω_n the form of $M_{\rm eff}(\omega)$ indeed corresponds to the presence of frequency-independent mass deficit deficit (as it was suggested by Andreev [6]). With the decrease in frequency the value of mass deficit decreases and passes through zero at frequencies at which the dissipative contribution is most prominent. At $\omega \to 0$ both parts (dissipative and nondissipative) of the TLS-induced correction to $1/M$ tend to zero.

5. The case of incoherent tunnelling. Consider now the case when the tunnelling process inside a two-level system is incoherent. In such a situation the displacement of the TLS with respect to the solid frame $\mathbf{u} \equiv \mathbf{x} - \mathbf{r}$ induced by a time-dependent external force $\mathbf{f}(t) \propto \cos(\omega t)$ coupled to the solid frame's position $\mathbf{r} = \mathbf{R} - (m/M)\mathbf{u}$ acquires a very simple form [9],

$$\mathbf{u}(\omega) = -\frac{1}{-i\tau\omega + 1}\frac{m}{M}\frac{\mathbf{a}(\mathbf{af})}{4T\cosh^2(\epsilon/T)}, \tag{19}$$

where relaxation time τ is inversely proportional to the tunnelling rate, whereas ε retains the same meaning as above.

Substitution of Eq. (19) into the classical analog of Eq. (9), namely

$$\frac{d}{dt}\mathbf{v} = \frac{1}{M}\left[\mathbf{f} - \frac{m}{M}\frac{d}{dt}\frac{d\mathbf{u}}{\mathbf{dt}}\right], \tag{20}$$

then leads to the following expression for the frequency-dependent coefficient of proportionality in the relation $(d/dt)\mathbf{v} = \mathbf{f}/M_{\rm eff}$:

$$\frac{1}{M_{\rm eff}(\omega)} = \frac{1}{M} - \frac{\omega^2}{12TM^2}\sum_n \frac{1}{-i\tau_n\omega + 1}\frac{m_n^2 a_n^2}{\cosh^2(\varepsilon_n/T)}. \tag{21}$$

The form of Eq. (21) demonstrates that in the case of incoherent tunnelling the TLSs induced contribution to the effective mass can never be described in the form of frequency independent mass deficit and at high frequencies is of the dissipative nature.

6. Conclusion. In the present note we have analyzed how the presence of TLSs influences the dynamic properties of a solid and have demonstrated that in order to observe a reduction of the effective mass of the sample (analogous to that in superfluids) the solid should incorporate quantum TLSs and the frequency at which the external force is applied should be high enough in comparison with resonance times of the TLSs. In the case of incoherent tunnelling the regime in which the contribution from the TLSs can be described as frequency-independent effective mass deficit is absent. Since TLSs are local objects, analogous conclusions are applicable also to the effective moment of inertia $I_{\rm eff}(\omega)$, which is a relevant quantity in torsional oscillator experiments. Experimentally, the crossover to the regime which can be associated with the appearance of an effective mass deficit has been demonstrated in a number of works starting from those of Kim and Chan [1]. The detailed comparison of the temperature dependences of both components of the response can be found in Ref. [10], which also contains a comprehensive list of references to other experimental and theoretical works.

An essential feature of our results is that in both regimes (of coherent and incoherent tunnelling) the TLS-induced contribution vanishes in the limit of $\omega \to 0$. Another common feature of the two cases is that the presence of the TLSs makes an additive contribution to $1/M_{\rm eff}$ or $1/I_{\rm eff}$ rather than to $M_{\rm eff}$ or $I_{\rm eff}$. In that respect the situation is quite analogous to that in superconducting vortex glasses where TLSs make a positive contribution to the inverse superfluid density (in other terms, specific inductance) rather then a negative contribution to the superfluid density itself [9, 11].

It seems worthwhile to mention that the phenomenological approach [12] used in Ref. [10] for analyzing the experimental data is based on conjectures which are in contradiction with both these properties. Namely, the authors of Ref. [12] have assumed that the internal degrees of freedom of solid ^{4}He make a contribution to its back action which in terms of the frequency-dependent effective moment of inertia of He sample, $I_{\rm eff}(\omega)$, can be written as an additive correction to the frequency-independent moment of inertia of the solid frame,

$$I_{\rm eff}(\omega) = I_0 + \frac{g_0}{\omega^2(1-i\tau\omega)^\beta} \qquad (22)$$

with $\beta \leq 1$. Moreover, at low frequencies this contribution does not tend to zero but behaves itself like a *negative* correction to the stiffness constant of the torsional oscillator. If it were really so, the solid by itself (outside of the torsional oscillator) would be unstable. A more logical assumption on the form of $I_{\rm eff}(\omega)$ in a glassy system with incoherent tunnelling inside the TLSs [consistent with the form of Eq. (21)] would be

$$I_{\rm eff}(\omega) = \left[I_0^{-1} - \frac{\bar{g}\omega^2}{(1-i\tau\omega)^\beta}\right]^{-1}. \qquad (23)$$

The main argument of Ref. [10] in favor of the superglass state consists in the impossibility to reconcile experimental data with dependence (22) – the observed frequency shift is too large in comparison with the maximum in dissipation (characterized by the inverse quality factor), the same being true also for dependence (23). However, the dispersion of the parameters of the TLSs can be taken into account by the replacement of $\beta = 1$ by $\beta < 1$ only for a particular form of the distribution and in other cases may lead to different dependences, especially if the appearance of the frequency shift is induced by the crossover between the regimes of incoherent and coherent tunnelling inside the TLSs. To elucidate this point the experimental investigations of the temperature dependence of the solid ^{4}He dynamic response have to be complemented by more systematic studies of its frequency dependence.

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