Tunnelling defect nanoclusters in hcp $^4$He crystals: alternative to supersolidity

A.F. Andreev

Kapitza Institute for Physical Problems, Russian Academy of Sciences, Kosygin Str. 2, Moscow, 119334 Russia

Abstract

A simple model based on the concept of resonant tunnelling clusters of lattice defects is used to explain the low temperature anomalies of hcp $^4$He crystals (mass decoupling from a torsional oscillator, shear modulus anomaly, dissipation peaks, heat capacity peak). Mass decoupling is a result of an internal Josephson effect: mass supercurrent in phase coherent tunnelling clusters. Quantitative results are in reasonable agreement with experiments.

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1 Introduction

Helium crystals are the most pronounced examples of the quantum crystals in which quantum tunnelling of particles results in many unusual phenomena. Delocalization of impurities, vacancies, dislocations, and surface defects leads to quantum diffusion, vacancy-induced mobility of impurities and ions, internal friction anomalies, and weakly decaying crystallization waves, respectively (see the review paper [1]).

Motivated by theoretical predictions [2, 3, 4] of the superfluidity of solid $^4$He, Kim and Chan (KC) performed experiments [5, 6] similar to the Andronikashvili experiment [7, 8] in which the decoupling of the superfluid fraction of liquid HeII from a torsional oscillator (TO) was discovered. The remarkable observation of KC was a similar decoupling of a part of solid helium from TO below 0.2 K which was interpreted as the superfluidity of a solid.

However, unlike the superfluid transition, the onset of the mass decoupling is broad and is accompanied by a dissipation peak. Day and Beamish [9] measured the elastic shear modulus, and observed a similar behavior (stiffening and

*E-mail: andreev@kapitza.ras.ru
dissipation) in the same temperature range. Near the decoupling onset temperature, a broad heat capacity peak was observed \[10\], but no pressure-induced superflow through the solid was found \[11, 12\]. The magnitudes of anomalies depend strongly on the way the solid was prepared \[13\]. All these anomalies seem to be absent in perfect crystals.

In this paper (see also earlier letter \[14\]), a simple model is proposed to explain the low-temperature anomalies of imperfect \(^4\)He crystals. As in papers \[15, 16\], the concept of tunnelling two-level systems (TLSs) in solids \[17\] is used. In \[15, 16\] TLSs were considered in highly disordered (glassy) samples. In this case, the parameters of TLSs are uniformly distributed according to the original tunnelling model \[18\]. However, in the most experiments, solid \(^4\)He samples were grown by the blocked capillary technique, but they consisted of poly-crystals. TLSs in crystals are degenerate (or resonant) TLSs in which the bare (with no tunnelling) energy difference of two localized states is zero (see \[17\] and below where a simple example is presented). The physical reason of the degeneracy is the crystal symmetry. Two localized states transform to each other under a crystal symmetry transformation. (Otherwise, there is no reason for energy difference to be small in crystals). The degenerate TLS is only the simplest case. Tunnelling systems may generally consist of more than two localized configurations \[17\].

In our simple model, we suppose that the main contribution to all anomalies is introduced by degenerate TLSs of a certain (or crystallographically equivalent) structure. In this case, the characteristic parameters, including tunneling amplitudes, are the same for all TLSs.

The key point is a peculiar quantum phenomenon of momentum deficit for TLSs in moving solids \[15, 16\]. In a solid with local velocity \(v\), the momentum of a TLS can under certain conditions (see below) be equal to \(m^*v\), where \(m^*\) is the effective mass which is different from the contribution \(m\) of the TLS to the total mass. Generally, \(m^*\) depends on the frequency and amplitude of the local velocity oscillations, the difference \(m^* - m\) being always negative. Therefore, the TLS has nonzero internal momentum \((m^* - m)v\) directed opposite to the velocity of the solid. In classical physics, a system of particles moving in a restricted spatial region, has zero internal momentum up to the frequencies on the order of the characteristic frequency of particle oscillations. For TLSs, this frequency is the tunneling frequency. We show below that under conditions of KC experiments, TLSs in solid helium have the frequency independent internal momentum down to the frequencies six orders of magnitude lower than the tunneling frequency. The internal momentum disappears at frequencies below the reciprocal transverse relaxation time (the TLS phase memory time).

2 TLS in imperfect crystals

Simple example of a resonant TLS is a four-vacancy cluster in a hcp \(^4\)He crystal (see Fig.1). Vacancies are located in the apexes of a tetrahedron. Three of them are disposed in the symmetry plane, which is perpendicular to the c-axis of the
crystal. The fourth vacancy occupies one of the two positions which transform to each other by the reflection in the plane. According to numerical calculations [19], four-vacancy clusters in solid helium are either unbound or are bound too weakly at the temperatures of KC-experiments. However, highly metastable growth-introduced vacancy clusters can persist in crystals for a long time. The same can be true for clusters of a different metastable phase in hcp-crystals [20]. Large growth-introduced vacancy clusters are probably the faceted liquid bubbles studied experimentally in $^4$He crystals [21].

The Hamiltonian of a TLS in a stationary crystal is

$$H_0 = \xi \sigma_3 + \Delta \sigma_1,$$

where $\sigma_\alpha$, ($\alpha = 1, 2, 3$), are the Pauli matrices, $\Delta$ is the tunneling amplitude, and $\mp \xi$ ($\xi > 0$) are the bare energies of two localized states. Here, the TLS is supposed to be nondegenerate, because we consider below a shear deformation that breaks the symmetry.

### 3 TLS in rotating crystals

The Hamiltonian of the TLS in a crystal moving with a local velocity $\mathbf{v}$, according to Galilean transformation is $H_1 = H_0 + \mathbf{p} \mathbf{v}$, where $\mathbf{p}$ is the TLS momentum operator. We suppose that the TLS tunneling is accompanied by the displacement of a mass $m$ by a vector $\mathbf{a}$. The operator part of the TLS center of gravity coordinate can be written as $r = -\sigma_2 \mathbf{a}/2$. The momentum operator is $\mathbf{p} = m \dot{r} = (im/\hbar)[H_0, r] = -(m\Delta/\hbar)\sigma_2$ [15][16].

The internal momentum in the TLS state $\psi_1 |1\rangle + \psi_2 |2\rangle$ is proportional to $\langle \sigma_2 \rangle \propto \sin(\varphi_1 - \varphi_2)$. Here, $|1\rangle$ and $|2\rangle$ are the localized states, and $\varphi_1$ and $\varphi_2$ are the phases of $\psi_1$ and $\psi_2$, respectively. Therefore, the internal momentum is caused by a kind of internal Josephson effect: the mass supercurrent inside phase coherent TLSs.
We suppose that the velocity is a result of an axisymmetric container rotation. Otherwise, additional terms should be added to the Hamiltonian to take the macroscopic displacement of the container walls into account (see [22], §11). Because the size of the TLS is assumed to be much smaller than the length scale of the rotating container, we can use the following expressions for the velocity and the TLS angular momentum: \( \mathbf{v} = \Omega \times \mathbf{R}, \) and \( M = (\mathbf{R} \times \mathbf{p})_z, \) where \( \Omega \) is the angular velocity and \( \mathbf{R} \) is the coordinate of the TLS (\( \mathbf{R} \perp \Omega \)) with respect to the origin located at the rotation axis which is parallel to the z-axis. We have

\[
H_1 = H_0 + M\Omega,
\]

where \( M = \mu\sigma_2, \) \( \mu = -(m\Delta/\hbar)aR\cos \theta, \) \( \theta \) is the angle between \( \mathbf{a} \) and \( \mathbf{v}. \)

After canonical transformation by the unitary operator

\[
U = (2\varepsilon(\varepsilon + \xi))^{-1/2}(\varepsilon + \xi - i\Delta \sigma_2),
\]

where \( \varepsilon = (\Delta^2 + \xi^2)^{1/2}, \) we obtain the Hamiltonian in the more convenient form

\[
H = UH_1U^+ = -\varepsilon\sigma_3 + \mu\Omega\sigma_2.
\]

This Hamiltonian can be written in the form

\[
H = -h_\alpha\sigma_\alpha,
\]

where \( h_\alpha \) is the “field” with components \( h_1 = 0, \) \( h_2 = -\mu\Omega, \) and \( h_3 = \varepsilon. \)

The TLS density matrix \( \rho \) is generally determined by the real polarization vector \( s_\alpha: \)

\[
\rho = (1 + s_\alpha\sigma_\alpha)/2.
\]

We have

\[
\langle \sigma_\alpha \rangle = \text{Tr} (\rho\sigma_\alpha) = s_\alpha.
\]

From the equation for the density matrix

\[
\dot{\rho} = (i/\hbar)[\rho, H],
\]

we obtain the dynamic equation for a free TLS (without dissipation):

\[
\hbar\dot{s}_\alpha = e_{\alpha\beta\gamma}h_\beta s_\gamma,
\]

where \( e_{\alpha\beta\gamma} \) is the Levi-Civita tensor.

The adiabatic theorem (see [23], chap II, §5c) takes place as a consequence of (8): along with the modulus \( s = |s_\alpha| \) of polarization, the angle between the field \( h_\alpha \) and \( s_\alpha \) is the integral of motion. The process is adiabatic if the time scale of the field variation is much longer than \( \hbar/|h_\alpha| \). The last condition is fulfilled with a significant safety margin: the characteristic frequency of the field variation in experiments mentioned above, is on the order of 1 kHz, but the magnitude of the field \( |h_\alpha| \) is on the order of \( \Delta \sim 0.1 \text{ K}. \)

Until the rotation (and deformation) is applied, the polarization is directed along the field, and the absolute value of the equilibrium polarization is \( s = s_0, \)
where $s_0 = \tanh \varepsilon/T$. According to the adiabatic theorem, we have for the free TLS:

$$s_1 = 0, \quad s_2 = -\frac{\mu}{E}\Omega s, \quad s_3 = \frac{\varepsilon}{E} = s - \frac{\mu^2\Omega^2}{E(E + \varepsilon)}s,$$

(9)

where $E = (\varepsilon^2 + \mu^2\Omega^2)^{1/2}$ is the field modulus.

We suppose that equations describing relaxation processes in a stationary solid have the standard form:

$$\dot{s}_2 = -\frac{s_2}{\tau_2}, \quad \dot{s}_3 = -\frac{s_3 - s_0}{\tau_1},$$

(10)

where $\tau_1$ and $\tau_2$ are the longitudinal and transverse relaxation times, respectively. Additional terms to the time derivatives (10) taking the rotation of the solid into account, can be determined from (9). Finally, we obtain the following dynamic equations for the TLS:

$$\frac{\partial}{\partial t} \left( s_2 + \frac{\mu\Omega}{E}s \right) = -\frac{s_2}{\tau_2}, \quad \frac{\partial}{\partial t} \left( s_3 + \frac{\mu^2\Omega^2}{E(E + \varepsilon)}s \right) = -\frac{s_3 - s_0}{\tau_1}.$$

(11)

Following the work of Burin et al. [24], we will assume that both reciprocal relaxation times depend linearly on temperature:

$$\tau_1^{-1} = \chi_1 T, \quad \tau_2^{-1} = \chi_2 T,$$

(12)

where $\chi_1$ and $\chi_2$ are constants.

4 Torsional oscillations

Consider torsional oscillations with a small amplitude $\Omega(t) \propto \exp(-i\omega t)$. From the first of the equations (11), we obtain the mean value of the TLS angular momentum $\langle M \rangle = \mu s_2 = I(\omega)\Omega$, where

$$I(\omega) = \frac{\mu^2 i\omega \tau_2 - \omega^2 \tau_2^2}{\Delta} \frac{\omega^2 \tau_2^2 + 1}{\omega^2 \tau_2^2 + 1} \tanh \frac{\Delta}{T},$$

(13)

is the TLS rotational inertia. The real part $I'(\omega)$ of $I$ determines the TLS mass deficit $\delta m(\omega) = m - m^* = -I'(\omega)/R^2$. The crystal fraction $f$, decoupled from TO, is $f = n\delta m/\rho$, where $\rho$ is the crystal density, and $n$ is the number of resonant TLSs per unit volume. We have

$$\delta m(\omega) = \left( \frac{ma \cos \theta}{h} \right)^2 \frac{\omega^2 \tau_2^2}{\omega^2 \tau_2^2 + 1} \Delta \tanh \frac{\Delta}{T}.$$

(14)

The mass deficit of a free TLS ($\omega \tau_2 \gg 1$) is frequency independent and coincides with the result of [16].

For uniform ($\omega = 0$) rotation $\delta m = 0$, in accordance with [15, 16]. The intermediate “quasi-equilibrium” region which was considered in [15, 16], is absent in our simple model.
Korshunov [25] calculated the TLS mass deficit for translational oscillations of a closed container filled with solid helium. His value of $\delta m$ is proportional to the extremely small factor $(\omega/\Delta)^2$. In fact, the Korshunov result is an explanation of the absence of TO anomalies in the blocked annulus experiments [6, 26].

The contribution $\langle \dot{E} \rangle = \langle \Omega \dot{M} \rangle$ of the TLS to the energy dissipation is determined by the imaginary part $I''(\omega)$ of $I$ (see [22, §123]): $\langle \dot{E} \rangle = (\omega/2)I''(\omega)|\Omega|^2$.

Under experimental conditions [27], the dependence of the reciprocal quality factor of TO on temperature and frequency, is determined by the expression

$$Q^{-1} \propto \frac{\omega v^2}{\omega^2 v^2 + 1} \tanh \frac{\Delta}{T}.$$  

(15)

The temperature dependence of (15) for two different frequencies $\omega$ are shown at the bottom of Fig.2. Note that according to (15), these curves intersect at some point.

The corresponding formula for the decoupled fraction $f$, according to (14), is

$$f \propto \frac{\omega^2 v^2_0}{\omega^2 v^2_0 + 1} \tanh \frac{\Delta}{T}.$$  

(16)

The temperature dependencies of (16) for the same two frequencies are shown at the top of Fig.2.

5 Nonlinear torsional oscillations

Consider nonlinear torsional oscillations at low temperatures (free TLSs). The oscillations are described by the equation

$$\ddot{\alpha} + \omega_0^2 \alpha = -(1/I_0)\dot{M}(t)$$  

(17)

for the rotation angle $\alpha$ ($\dot{\alpha} = \Omega$). Here, $I_0$ and $\omega_0$ are the rotational inertia and the TO fundamental frequency in the absence of TLSs, respectively. $M(t)$ is the TLS angular momentum. According to (1) and (9) we have

$$M(t) = -\frac{R\Delta}{v_c^2} \frac{v \cos^2 \theta}{(1 + \cos^2 \theta(v/v_c)^2)^{1/2}},$$  

(18)

where $v_c = \hbar/(ma)$ is a characteristic (critical) velocity.

In the absence of TLSs, the oscillations are harmonic: $\alpha(t) = \alpha_0 \cos \varphi$, where $\alpha_0$ is the amplitude and $\varphi = \omega_0 t$. The time dependence of $M$ is determined by (18) with $v = v_0 \sin \varphi$, $v_0 = -\alpha_0 \omega_0 R$. Being the periodic function of $\varphi$, $M(t)$ can be represented by its Fourier series. It is known [22] that a small correction to the fundamental frequency is determined by the resonant part (proportional to $\cos \varphi$) of the right hand side of (14). Resonant contribution to the momentum $M_{\text{res}}(\varphi)$ is proportional to $\sin \varphi$

$$M_{\text{res}}(\varphi) = \sin \varphi \frac{\hbar}{\pi} \int_{-\pi}^{\pi} M(\varphi') \sin \varphi' d\varphi'.$$  

(19)
Figure 2: Temperature dependencies of decoupled fraction $f$ and dissipation $\chi_2$ ($\Delta = 51$ mK, $\chi_2 = 5.9$ kHz/K) for two frequencies fitting experimental data [27].
According to (18) and (19), the TLS contribution to the fundamental frequency and, therefore, to the value of the decoupled fraction \( f(v_0) \) is determined by the expression

\[
 f(v_0) = \text{const} \cos^2 \theta \int_{-\pi}^{\pi} \frac{\sin^2 \varphi}{(1 + \cos^2 \theta (v_0/v_c)^2 \sin^2 \varphi)^{1/2}}. \tag{20}
\]

We consider a narrow annulus where \( R \approx \text{const} \) for all TLSs. For poly-crystals, the expression (20) should be averaged over all directions of \( \mathbf{a} \) at a given \( v \). We have

\[
 f(v) = \text{const} \int_{-1}^{1} \cos^2 \theta d \cos \theta \int_{-\pi}^{\pi} \frac{\sin^2 \varphi d\varphi}{(1 + (v/v_c)^2 \cos^2 \theta \sin^2 \varphi)^{1/2}}. \tag{21}
\]

Here and below we omit the index 0 in the velocity amplitude \( v_0 \). Finally, we obtain

\[
 f(v)/f(0) = \frac{12}{\pi} \int_0^1 x^2 dx \int_0^{\pi/2} \frac{\sin^2 \varphi d\varphi}{(1 + (v/v_c)^2 x^2 \sin^2 \varphi)^{1/2}}. \tag{22}
\]

This dependence is plotted in Fig.3. According to (14), the mass deficit \( \delta m \) for a single TLS is on the order of \( \Delta^2/v_c^2 \). Assuming \( \Delta \sim 0.1 \text{ K} \) and \( v_c \sim 10 \mu \text{m/s} \), we obtain \( \delta m \sim 10^{-11} \text{ g} \). The experimental value of the decoupled fraction \( f = n\delta m/\rho \) corresponds to the quite low TLS concentration \( n \sim 10^7 \text{ cm}^{-3} \).

The characteristic size \( L \) of the defect clusters can be estimated from the value of the critical velocity. Assuming that \( ma = \rho L^4 \), we have \( L \sim 10 \text{ nm} \).

### 6 The shear modulus anomaly

Let \( \zeta = \partial u_x/\partial y \) be a shear strain (\( u_x \) is the displacement along the \( x \)-axis). According to (3) in the absence of rotation the derivative of the TLS Hamiltonian with respect to the strain is \( \partial H/\partial \zeta = -\sigma_3 \partial \varepsilon/\partial \zeta \). The TLS contribution to the stress \( \sigma \) is determined by the mean value of this derivative

\[
 \sigma = n < \partial H/\partial \zeta > = -n s_3 \partial \varepsilon/\partial \zeta. \tag{23}
\]

We consider small oscillations of the strain \( \zeta(t) \propto \exp(-i\omega t) \). The oscillating part of the TLS polarization \( s'_3 \) and that of the instantaneous equilibrium polarization \( s'_0 \) satisfy, according to (10), the equation \( i\omega s'_3 = (s'_3 - s'_0)/\tau_1 \), where

\[
 s'_0 = \zeta \frac{\partial}{\partial \zeta} \tanh \frac{\varepsilon}{T} = \frac{\zeta}{T \cosh^2(\Delta/T)} \left( \frac{\partial \varepsilon}{\partial \zeta} \right)_0. \tag{24}
\]

Here, \( (\partial \varepsilon/\partial \zeta)_0 \) is the derivative \( \partial \varepsilon/\partial \zeta \) at \( \zeta = 0 \). We have

\[
 s'_3 = \frac{1}{1 - i\omega \tau_1} \frac{\zeta}{T \cosh^2(\Delta/T)} \left( \frac{\partial \varepsilon}{\partial \zeta} \right)_0. \tag{25}
\]

The oscillating part of the stress is

\[
 \sigma' = -n \left\{ \zeta \left( \frac{\partial^2 \varepsilon}{\partial \zeta^2} \right)_0 \tanh \frac{\Delta}{T} + \left( \frac{\partial \varepsilon}{\partial \zeta} \right)_0 s'_3 \right\}. \tag{26}
\]
Figure 3: Normalized decoupled fraction \( f(v) = \frac{v}{f(0)} \) versus oscillation velocity amplitude fitting experimental data \[29\].

Figure 4: Experimentally observed \[30\] temperature dependence of shear modulus at high frequency and a fit for \( \Delta = 0.08 \text{ K} \).
The TLS contribution to the complex shear modulus $G(\omega)$ defined by the expression $\sigma' = G(\omega)\zeta$, is

$$G(\omega) = n \left\{ \left| \frac{\partial^2 \varepsilon}{\partial \zeta^2} \right| \tanh \frac{\Delta}{T} - \frac{1}{1 - i\omega\tau} \frac{1}{T \cosh^2(\Delta/T)} \left( \frac{\partial \varepsilon}{\partial \zeta} \right)^2 \right\}. \quad (27)$$

The second derivative $\partial^2 \varepsilon/\partial \zeta^2$ should be negative. Otherwise, the real part of $G$, contrary to experiments [9], would be negative.

The dissipation of the shear oscillations is determined by the imaginary part of $G$. Its temperature dependence is characterized by the presence of a characteristic peak, in accordance with experiments [9].

It was noted [9] that $G'$ and $f$ change very similarly with temperature. This is consistent with our results: due to (16) and (27), in high frequency region $\omega\tau \gg 1$, we have $G \propto f \propto \tanh(\Delta/T)$. This dependence for $G$ is plotted in Fig.4.

### 7 Thermodynamics of resonant TLSs

The contribution of resonant TLSs to the free energy of the unit volume of a crystal is

$$F = -nT \log \left( 2 \cosh \frac{\Delta}{T} \right). \quad (28)$$
The heat capacity is determined by the well-known formula:

\[ C = -T \frac{\partial^2 F}{\partial T^2} = n \left( \frac{\Delta}{T} \right)^2 \cosh^{-2} \frac{\Delta}{T}. \]  

(29)

This dependence is plotted in Fig.5.

The contribution of TLSs to pressure is

\[ P = \rho \left( \frac{\partial F}{\partial \rho} \right)_T = -n \rho \frac{\partial \Delta}{\partial \rho} \tanh \frac{\Delta}{T}. \]  

(30)

8 Summary

Low-temperature anomalies of hcp $^4$He crystals are caused by growth-introduced resonant nanoscale clusters of lattice defects. The quantum phase coherence and tunnelling of the clusters are crucial. The important problem is to identify the structure of the clusters. The most probable candidates seem to be the faceted liquid bubbles.

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