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Low-temperature anomalies of ^{4}He crystals

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Low-temperature anomalies of hcp ^{4}He crystals (mass decoupling from a torsional oscillator, shear modulus anomaly, dissipation peaks, heat capacity peak) are explained. A simple model based on the concept of resonant tunneling systems in imperfect crystals is proposed. Mass decoupling is caused by an internal Josephson effect: the mass supercurrent inside phase coherent tunneling systems. Quantitative results are in reasonable agreement with experiments.

1. Introduction. Kim and Chan (KC), motivated by predictions [1–3] of the superfluidity of solid ^{4}He, performed experiments [4, 5] similar to famous Andronikashvili experiment [6, 7] 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 et al. [8] measured the elastic shear modulus, and observed a similar behavior (stiffening and dissipation) in the same temperature range. Near the decoupling onset temperature, a broad heat capacity peak was observed [9], but no pressure-induced superflow through the solid was found [10, 11]. The magnitudes of anomalies depend strongly on the way the solid was prepared [12]. All these anomalies seem to be absent in perfect crystals.

In this letter, a simple model is proposed to explain the low-temperature anomalies of imperfect ^{4}He crystals. As in earlier papers [13, 14], the concept of tunneling two-level systems (TLSs) in solids [15] is used. In [13, 14] TLSs were considered in highly disordered (glassy) samples. In this case, the parameters of TLSs are uniformly distributed according to the original tunneling model [16]. However, in most experiments, solid ^{4}He samples were grown by the blocked capillary technique, but they consisted either of single crystals or polycrystals. TLSs in crystals are degenerate TLSs in which the bare (with no tunneling) energy difference of two localized states is zero (see [15] 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. Tunneling systems may generally consist of more than two localized configurations [15].

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 the momentum deficit for TLSs in moving solids [13, 14]. In a solid with local velocity $\mathbf{v}$, the momentum of a TLS can under certain conditions (see below) be equal to $m^*\mathbf{v}$, 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 $(m^* - m)\mathbf{v}$ 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. TLSs in imperfect crystals. Simple example of a resonant TLS is a four-vacancy cluster in a hcp ^{4}He crystal. 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 [17], four-vacancy clusters in solid helium are either unbound

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or are bound too weakly at the temperatures of KC-experiments. However, highly metastable small growth-introduced vacancy clusters can persist in crystals for a long time. The same can be true for small clusters of a different metastable phase in hcp crystals [18]. Resonant TLSs can also appear when a defect cluster resides on symmetric extensive defects like stacking faults.

The Hamiltonian of a TLS in a stationary crystal is $H_0 = \xi\sigma_3 + \Delta\sigma_1$, where σ_α ($\alpha = 1,2,3$) are the Pauli matrices, Δ 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. TLSs in rotating crystals. The Hamiltonian of the TLS in a crystal moving with a local velocity $\mathbf{v}$, according to Galilean transformation is $H_1 = H_0 + \mathbf{pv}$, 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 $\mathbf{r} = -\sigma_3\mathbf{a}/2$. The momentum operator is $\mathbf{p} = m\dot{\mathbf{r}} = (im/\hbar)[H_0\mathbf{r}] = -(m\Delta/\hbar)\mathbf{a}\sigma_2$ [13, 14].

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 φ_1 and φ_2 are the phases of ψ_1 and ψ_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 axially symmetric container rotation. Otherwise, additional terms should be added to the Hamiltonian to take the macroscopic displacement of the container walls into account (see [19], §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 Ω is the angular velocity and $\mathbf{R} \perp \Omega$ is the coordinate of the TLS 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 $M = \gamma\sigma_2$,

$$\gamma = (m\Delta/\hbar)(\mathbf{a}_\perp \times \mathbf{R})_z = (m\Delta/\hbar)a_\perp R \sin\theta, \tag{1}$$

$\mathbf{a}_\perp$ is the projection of $\mathbf{a}$ to the (xy)-plane, and θ is the angle between $\mathbf{a}_\perp$ and $\mathbf{R}$.

After canonical transformation by the unitary operator

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

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

$$H = UH_1U^+ = -\varepsilon\sigma_3 + \gamma\Omega\sigma_2. \tag{3}$$

This Hamiltonian can be written in the form

$$H = -h_\alpha\sigma_\alpha, \tag{4}$$

where h_α is the "field" with components $h_1 = 0$, $h_2 = -\gamma\Omega$, and $h_3 = \varepsilon$.

The TLS density matrix w is generally determined by the real polarization vector s_α

$$w = (1 + s_\alpha\sigma_\alpha)/2. \tag{5}$$

We have

$$\langle\sigma_\alpha\rangle = \mathrm{Tr}\,(w\sigma_\alpha) = s_\alpha. \tag{6}$$

From the equation for the density matrix

$$\dot{w} = (i/\hbar)[w, H], \tag{7}$$

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

$$\hbar\dot{s}_\alpha = e_{\alpha\beta\gamma}h_\beta s_\gamma, \tag{8}$$

where $e_{\alpha\beta\gamma}$ is the Levi–Civita tensor.

The adiabatic theorem (see [20], 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_α and s_α 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 $\Delta \sim 0.1$ 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:

$$s_1 = 0, \quad s_2 = -\frac{\gamma}{E}\Omega s, \quad s_3 = \frac{\varepsilon}{E}s = s - \frac{\gamma^2\Omega^2}{E(E+\varepsilon)}s, \tag{9}$$

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

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

$$\dot{s_2} = -\frac{s_2}{\tau_2}, \quad \dot{s_3} = -\frac{s_3 - s_0}{\tau_1}, \tag{10}$$

where τ_1 and τ_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:

$$\frac{\partial}{\partial t}\left(s_2 + \frac{\gamma\Omega}{E}s\right) = -\frac{s_2}{\tau_2},$$
$$\frac{\partial}{\partial t}\left(s_3 + \frac{\gamma^2\Omega^2}{E(E+\varepsilon)}s\right) = -\frac{s_3 - s_0}{\tau_1}. \quad (11)$$

Following the work of Burin et al.[21], we will assume that both reciprocal relaxation times depend linearly on temperature:

$$\tau_1^{-1} = \chi_1 T, \quad \tau_2^{-1} = \chi_2 T, \quad (12)$$

where χ_1 and χ_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 $\langle M\rangle = \gamma s_2 = I(\omega)\Omega$, where

$$I(\omega) = \frac{\gamma^2}{\Delta}\frac{i\omega\tau_2 - \omega^2\tau_2^2}{\omega^2\tau_2^2 + 1}\tanh\frac{\Delta}{T} \quad (13)$$

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 $f = (N/\rho)\delta m$, where ρ is the crystal density, and N is the number of resonant TLSs per unit volume. We have

$$\delta m(\omega) = \left(\frac{ma_\perp \sin\theta}{\hbar}\right)^2 \frac{\omega^2\tau_2^2}{\omega^2\tau_2^2 + 1}\Delta\tanh\frac{\Delta}{T}. \quad (14)$$

The mass deficit of a free TLS ($\omega\tau_2 \gg 1$) is frequency independent and coincides with the result of [14].

For uniform ($\omega = 0$) rotation $\delta m = 0$, in accordance with [13, 14]. The intermediate "quasi-equilibrium" region which was considered in [13, 14], is absent in our simple model.

Korshunov [22] calculated the TLS mass deficit for translational oscillations of a closed container filled with solid helium. His value of δm 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 [5, 23].

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 [19], § 123): $\langle\dot{E}\rangle = (\omega/2)I''(\omega)\,|\Omega|^2$. Under experimental conditions [24], the dependence of the reciprocal quality factor of TO on temperature and frequency, is determined by the expression

$$Q^{-1} \propto \frac{\omega\tau_2}{\omega^2\tau_2^2 + 1}\tanh\frac{\Delta}{T}. \quad (15)$$

The temperature dependences of (15) for two different frequencies ω are shown at the bottom of Fig. 1. Note that according to (15), these curves intersect at some

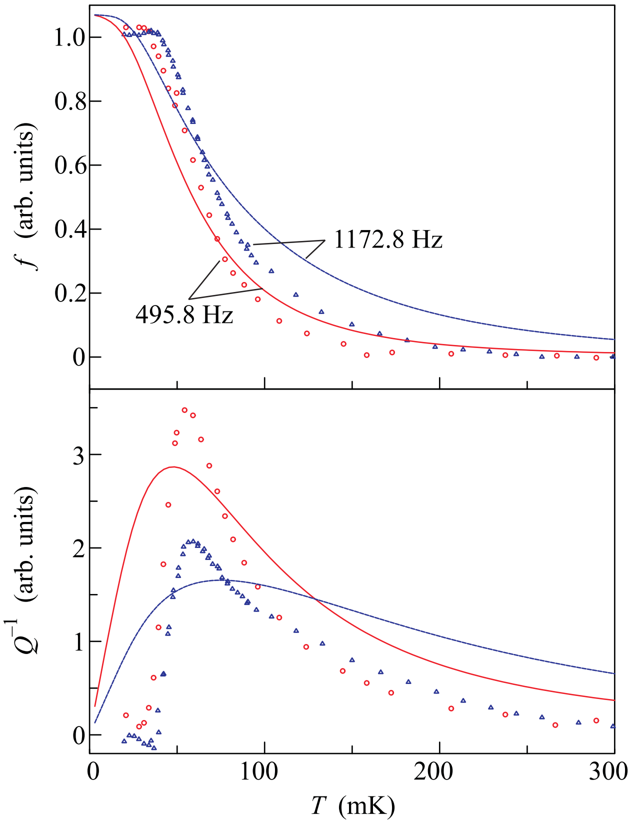

Fig. 1. Temperature dependences of decoupled fraction (16) and dissipation (15) (Δ = 51 mK, χ_2 = 5.9 kHz/K) for two frequencies fitting experimental data [24]

point.

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

$$f \propto \frac{\omega^2\tau_2^2}{\omega^2\tau_2^2 + 1}\tanh\frac{\Delta}{T}. \quad (16)$$

The temperature dependences of (16) for the same two frequencies are shown at the top of Fig. 1.

5. 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 σ is determined by the mean value of this derivative

$$\sigma = N\left\langle\frac{\partial H}{\partial\zeta}\right\rangle = -Ns_3\frac{\partial\varepsilon}{\partial\zeta}. \quad (17)$$

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

$$s_0' = \zeta \frac{\partial}{\partial \zeta} \tanh \frac{\varepsilon}{T} = \frac{\zeta}{T \cosh^2(\Delta/T)} \left(\frac{\partial \varepsilon}{\partial \zeta}\right)_0 . \qquad (18)$$

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

$$s_3' = \frac{1}{1 - i\omega\tau_1} \frac{\zeta}{T \cosh^2(\Delta/T)} \left(\frac{\partial \varepsilon}{\partial \zeta}\right)_0 . \qquad (19)$$

The oscillating part of the stress is

$$\sigma' = -N \left[\zeta \left(\frac{\partial^2 \varepsilon}{\partial \zeta^2}\right)_0 \tanh \frac{\Delta}{T} + \left(\frac{\partial \varepsilon}{\partial \zeta}\right)_0 s_3'\right] . \qquad (20)$$

The TLS-contribution to the complex shear modulus $G(\omega)$ determined by the expression $\sigma' = G(\omega)\zeta$, is

$$G(\omega) = \\ = N \left[\left|\frac{\partial^2 \varepsilon}{\partial \zeta^2}\right| \tanh \frac{\Delta}{T} - \frac{1}{1 - i\omega\tau_1} \frac{1}{T \cosh^2(\Delta/T)} \left(\frac{\partial \varepsilon}{\partial \zeta}\right)_0^2\right] . \qquad (21)$$

The second derivative $\partial^2\varepsilon/\partial\zeta^2$ should be negative. Otherwise, the real part of G, contrary to experiments [8], 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 [8].

It was noted [8] that G' and f change very similar with temperature. This is consistent with our results: due to (16) and (21), in high frequency region $\omega\tau \gg 1$, we have $G \propto f \propto \tanh(\Delta/T)$.

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

$$F = -NT \log 2 \cosh \frac{\Delta}{T} . \qquad (22)$$

The heat capacity is determined by the well-known formula:

$$C = -T \frac{\partial^2 F}{\partial T^2} = N \left(\frac{\Delta}{T}\right)^2 \cosh^{-2} \frac{\Delta}{T} . \qquad (23)$$

This dependence is plotted in Fig. 2.

The contribution of TLSs to pressure is

$$P = \rho \left(\frac{\partial F}{\partial \rho}\right)_T = -N\rho \frac{\partial \Delta}{\partial \rho} \tanh \frac{\Delta}{T} . \qquad (24)$$

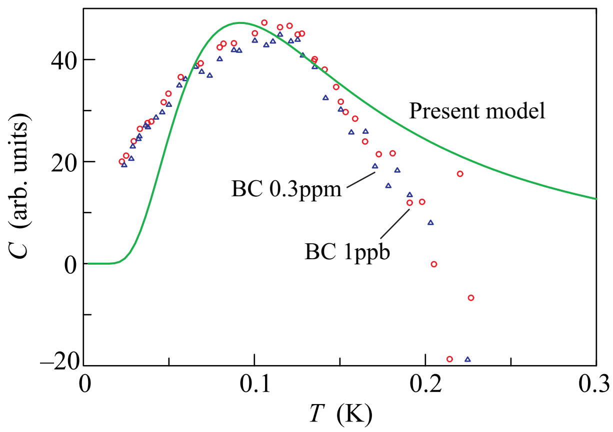

Fig. 2. Temperature dependence of heat capacity (23) ($\Delta = 0.11$ K) and experimental data [9]

$P(T)$ at a fixed volume was measured by Grigoriev et al. [25]. The glassy contribution ($\propto T^2$) was found. The term (24) was absent. The possible reason is that the derivative $\partial\Delta/\partial\rho$ is anomalously small. Indeed, this derivative determines in our model the pressure dependence of the onset temperature of the mass decoupling. KC [26] observed no apparent change of the onset temperature with pressure.

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