Spontaneous emergence of van der Waals interaction - a road to phase coherence at mK temperatures

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We present the experimental results on the spontaneous emergence of the phase coherence in the system of oscillating electric dipoles in piezo-resonators caused by the van der Waals interaction. Spontaneous emergence of the phase coherence in these systems is manifested via temperature-dependent, extremely accurate tune-up of their resonance frequencies in 9th order with relative spectral line-width $\delta f_\text{f}/f_\text{f}$ less than $3 \times 10^{-8}$ (this number is comparable with that in lasers). Moreover, we show that the application of an incoherent (noise) excitation signal leads to a spontaneous formation of the phase coherent state, and that the dissipation processes do not affect this phase coherent state (i.e. the resonance frequency of the system). All above-mentioned signatures are typical characteristics for a Bose-Einstein condensate of excitations.

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Bose-Einstein (B-E) condensation is a fundamental physical phenomenon when a macroscopic number of bosons condense into a collective quantum ground state governed by a single wave function. Text-book examples of the B-E condensates are superfluid $^4$He and ultracold atomic gases [1, 3]. In case of the fermions, the scenario of the B-E condensation is provided via a mutual coupling interaction between pair of fermions allowing them the formation of the bosonic Cooper pairs, and these pairs condense and occupy the single energy level that is by the energy gap $\Delta$ lower than the Fermi energy. Examples of such B-E condensates are superconductivity of electrons and superfluid $^3$He [4, 5]. Recently, however, a concept of the B-E condensation was extended also to the physical systems with a spontaneous emergence of the phase coherence [6]. These can be the systems of excitations, the lifetime of which is longer than the time they need to scatter and there is an interaction acting between them allowing to set a single energy state governed by the single wave function [7, 8]. Typical examples of such excitations involve magnons, excitons, polaritons, etc. [9–14]. Criterium of the spontaneous emergence of the phase coherence means that a physical system under consideration should exhibit a weak long-range interaction comparable with its thermal energy, and by cooling the system, this interaction (i) should overwhelm the thermal energy and (ii) should couple and adjust the excitations into a phase-coherent state. Dipole-dipole interactions, and in particular van der Waals interactions are one of the fundamental, but relatively weak interactions which could satisfy above mentioned criteria and they are present in many physical systems.

We focused our investigation on the presence of the van der Waals interactions in piezoelectric resonators in the form of commercially available quartz tuning forks resonating at nominal frequencies 32 kHz, 77 kHz and 100 kHz. These devices are intensively used in AFM and STM techniques [13, 17], physics of superfluids ($^3$He and $^4$He) [18–20], etc. It is generally known that when the standard quartz tuning forks oscillating in vacuum are cooled down to temperatures below $\sim 15$ K, these resonators undergo a transition into the high Q-value oscillation mode with the Q-value of the order of 10$^6$ or more [18, 21]. In order to measure quartz tuning fork with high Q-value, instead of using a traditional technique of the frequency sweep with continuous voltage excitation, we adapted and applied a pulsed-demodulation (P-D) technique (or heterodyning technique with pulsed excitation) [22]. P-D technique transforms the frequency of the measured signal to lower values without losing information about the original frequency and allows reduction of the sampling rate and increases the resolution of the frequency measurements. This allows to measure the decay signals from the freely oscillating resonators with high resolution in the frequency in 9th order. Free oscillations of the tuning forks at resonance frequency $f_{1f}$ generate an alternating piezoelectric current being detected by a custom-made current-to-voltage (I/V) converter in order to minimize the losses in detection circuit [23, 24]. The I/V converter’s output voltage signal is then measured by a lock-in amplifier operating as a demodulator.

Before their installation the tuning forks were removed from the original metal can and the former (magnetic) leads were replaced by copper wires having a diameter of 120 $\mu$m. These copper wires were electrically connected to tuning fork’s pads using a conductive silver epoxy and glued to a small piece of Stycast 1266 - epoxy impregnated paper in order to achieve a mechanical stiffness of the set-up. Cooling of the tuning forks was ensured by clamping these copper wires between two copper blocks and these blocks were screwed to the mixing chamber of the cryogen-free dilution refrigerator Triton 200. As an example, Fig. [1] shows the decay signal of 32 kHz tuning fork’s free oscillations measured in vacuum at temper-
FIG. 1: (Colour online) Free decay signal of the 32 kHz tuning fork’s oscillations measured using pulsed-demodulation technique. Insets: a time window showing fit (red line) to experimental data (points) using equation (1) (top), FFT of the presented signal showing the precision of the resonance frequency determination (bottom).

ature of 25 mK. The deposited energy (250 pulses with 5 mV\textsubscript{RMS} amplitude) used to excite the fork is equal to $33\times10^{-15}$ Joule. The tuning fork’s high Q-value is demonstrated by a long lasting decay signal of the order of several tens of seconds. Decay signals measured from 77 kHz and 100 kHz tuning forks had the same form, however, the duration of their decay signals in time was slightly shorter, up to 30 seconds for both 77 kHz and 100 kHz tuning forks. All measured decay signals were fitted using the expression

$$V(t) = V(0) e^{-\frac{t}{\tau}} \sin(2\pi f_{\text{sig}} t + \phi), \quad (1)$$

where fitting parameters $V(0)$ is the initial amplitude of the signal, $\tau$ is the relaxation time constant characterizing a damping process, $f_{\text{sig}}$ is the signal frequency and $\phi$ is the signal phase. Insets to Fig. 1 show a time window with the fit to experimental data using equation (1) and FFT spectrum of the signal. Fit applied to the data measured by P-D technique has the resolution in frequency measurement, $\delta f_{\text{sig}}$, determined by $\delta f_{\text{sig}} = f_{\text{sig}}/(f_{\text{samp}} T_s)$, while that using traditional FFT technique is $\delta f_{\text{sig}} = 1/T_s$, where $f_{\text{samp}}$ is the sampling frequency (1000 Hz) and $T_s$ is the signal duration in time. For the decay signal presented in Fig. 1 the fit to experimental data measured by P-D technique gives the resolution in frequency to be $\sim 30 \mu$Hz, while the resolution of traditional FFT technique is $\sim 10$ mHz.

Figure 2 shows measured temperature dependencies of the tuning forks’ resonance frequencies during warming and cooling sweeps of the dilution refrigerator in temperature range between 50 mK and 350 mK. Temperature of the mixing chamber was measured using SQUID noise thermometer MFFT-1 provided by Magnicon; the temperature calibration was cross-checked using home-made fixed-point device \cite{25}, and the rate of the temperature sweep was 1 mK per minute. We found that during warm up process the resonance frequencies of the tuning forks are decreasing. At constant temperature, the resonance frequencies stay constant and on subsequent cooling process, the resonance frequencies rise reproducibly again. It is worth to note that we were not able to measure the temperature of the tuning forks themselves and presented measurements are related to temperature of the mixing chamber. However, based on the reproducibility we assume that the tuning forks were in thermal equilibrium with mixing chamber in temperature range presented. Inset to Fig. 2 shows the time evolution of mixing chamber temperature. What could be a physical origin of the fine temperature dependence of tuning forks’ resonance frequencies? Below we present a simple phenomenological physical model.

In contrast to a classical mechanical oscillator, there are oscillating induced electric dipoles $\mathbf{p} = Q\mathbf{d} = \alpha(T)\mathbf{E}_{\text{loc}}$ ($Q$ is the charge and $\mathbf{d}$ is the distance between the charges that is proportional to deflection of the ions
from equilibrium position \(x\) in the tuning fork, and each of them experiences a local electric field with intensity \(\bar{E}_{\text{loc}}\) produced by other electric dipoles (and an initial voltage pulse). Once the electric dipole moment \(p\) is formed, the restoring force of magnitude

\[
F_{\text{loc}} \approx \frac{Q^2 x}{\alpha(T)}
\]

(2)

acts in order to bring together two charges of the dipole, where \(\alpha(T)\) is the temperature dependent polarizability and \(x\) is the deflection of the ion from equilibrium. We presume that at millikelvin temperature range this electric force \(F_{\text{loc}}\) contributes to the elastic restoring force, so the total restoring force magnitude can be expressed as

\[
F_{\text{res}} \approx (k + \frac{Q^2}{\alpha(T)}) x,
\]

(3)

where \(k\) is the spring constant. Polarizability \(\alpha(T)\) characterizes the competition between potential energy of the electric dipole \((-p \cdot \bar{E}_{\text{loc}} = -p E_{\text{loc}} \cos(\theta))\) acting to orient the dipole in direction of the electric field and thermal energy \((k_B T)\) having tendency to disorder this dipole order. Polarizability of the quartz excited by pulse can be expressed in form

\[
\alpha(T) = \frac{3\varepsilon_0}{N < \cos(\theta)>} = \frac{3\varepsilon_0}{N L(X)},
\]

(4)

where \(\varepsilon_0\) is the vacuum permittivity, \(N\) is the density of dipoles and \(< \cos(\theta)>\) is the mean value of \(\sum_i \cos(\theta)_i\), which is equal to the Langevin function \(< \cos(\theta)> = L(X) = \coth(X) - 1/X\) with \(X = p \cdot E_{\text{loc}}/(k_B T)\). Therefore, frequency of the tuning fork oscillations \(f_i\) is determined by two terms

\[
f_i(T)^2 = \frac{k}{4\pi^2 m_{\text{eff}}} + \frac{Q^2 N L(X)}{12\pi^2 \varepsilon_0 m_{\text{eff}}} = f_0^2 + f_1^2(T).
\]

(5)

While the first term, in presented model, corresponds to the temperature independent resonance frequency determined by elastic properties of the quartz, the second term reflects the temperature contribution to the resonance frequency due to ordering effect provided by van der Waals interaction acting between the dipoles.

Figure 3 shows the temperature dependencies of the tuning forks’ resonance frequencies normalized to the maximal values, measured in temperature range from 25 mK to \(\sim 1\) K. Solid lines represent the fits to experimental data using expression \(f_i = \sqrt{a + b L(T_c/T)}\) (see Eq. \(5\)), where \(a = f_0^2\), \(b\) is the fitting constant characterizing the additional contribution to stiffness due to the van der Waals interaction, \(T_c = p E_{\text{loc}}/k_B\) is the critical temperature and \(L(T_c/T)\) is above-mentioned Langevin function. Presented fits show a good qualitative agreement with the experimental data. In fact, the values of critical temperatures \(T_c\) for all three forks are almost the same: they are in range between 1.1 K and 1.5 K, which allows to estimate the energy of the van der Waals interaction to be of the order of \(\sim 2 \times 10^{-23}\) Joule (\(\sim 0.12\) meV). We should also note that \(b\) fitting constants grow linearly with inverse value of the effective mass of the forks that qualitatively supports presented model (see Eq. \(27\)).

Now, let us discuss the influence of intrinsic damping process on tuning forks’ resonance frequencies. As mentioned above, the intrinsic damping processes are reflected in the relaxation time constant \((\gamma_D \sim 1/\tau(T))\). Inset to Fig. \(3\) shows measured temperature dependencies of the relaxation time constants \(\tau\) for individual tuning forks. Dependencies presented in Fig. \(3\) clearly demonstrate contradiction with properties of the standard linear harmonic oscillators, where the resonance frequency depends on damping \((\omega^2 = \omega_0^2 - \gamma_D^2/4)\). Temperature dependencies of the resonance frequencies for measured forks behave independently on damping process, and this reveals that dissipation mechanisms are acting rather near the quartz surface than in its volume i.e. at the boundary, where the coherence is violated. Moreover, temperature dependencies of relaxation time constants \(\tau\) show “Schottky-like anomalies” i.e. the temperature extrema, which one can attribute to a presence of fine thermally activated dissipation mechanisms. As the surface of the quartz tuning forks is covered by the metal electrodes made of various elements (tin, silver, etc.), Schottky barriers are formed on this interface due to difference in energy spectra between metal electrodes and quartz. These Schottky barriers, as might-be ex-
ample, can open additional dissipation channels caused by thermally activated injection of electric charges from metal alloy to quartz. However, physical nature of the dissipation processes remain unclear up to now and an additional work needs to be done to elucidate this problem.

Temperature changes $\delta T$ and $\delta f$ in the order of $10^{-8}$K for all forks are of the same order of magnitude. Comparison of the calculated values of $\delta f$ with measured data suggests that tuning forks themselves are exposed to a heating effect or there are the temperature gradients present inside the forks leading to the frequency fluctuations, but sources of the heat or origin of the temperature gradients remain unknown yet.

In order to show the presence of spontaneous emergence of the phase coherence in the system of oscillating dipoles in quartz tuning forks, we tried to excite the fork’s oscillations using incoherent white noise signal, similar to the experiment performed with B-E condensate of magnons in superfluid $^3$He-B [29].

Observed temperature dependencies of the tuning forks’ resonance frequencies and its independence on damping processes indicate that the system of oscillating dipoles in quartz tuning forks preserves the phase rigidity i.e. the phase coherence due to van der Waals interaction, thus revealing the characteristics of a coherent state of the oscillating dipoles. In order to justify this assumption we performed several measurements of the temperature stability of the resonance frequencies of individual tuning forks at different but constant temperatures. Figure 4 presents the distributions of the tuning forks’ resonance frequencies measured at base temperature of $\approx 8$ mK. Lines show the Gaussian fits to experimental data.

![FIG. 4: (Colour online) Distributions of the tuning forks’ resonance frequencies measured by technique described above at constant temperature of $\approx 8$ mK. Lines show the Gaussian fits to experimental data.](image)

In conclusion - we presented the experimental results on the spontaneous emergence of the phase coherence in the system of oscillating electric dipoles in quartz piezo-resonators caused by the van der Waals interaction. Spontaneous emergence of the phase coherence in these systems is manifested via temperature-dependent, extremely accurate tune-up of their resonance frequencies in 9th order with relative spectral line-width $\delta f_0/f_0$.
less than $3.10^{-8}$. Moreover, we showed that the application of an incoherent (noise) excitation signal leads to a spontaneous formation of the phase coherent state and that dissipation processes do not affect this phase coherent state (i.e., the resonance frequency of the system). All above mentioned signatures are typical characteristics for a B-E condensate of excitations. Whether this phenomenon is universal for the broad class of piezoelectric materials, and what is the nature of the dissipation mechanisms, these are the questions that need to be answered.

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