Violation of the London Law and Onsager-Feynman quantization in multicomponent superconductors

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Two quite fundamental principles governing the response to rotation of single-component superfluids and superconductors, the London law \cite{London} relating the angular velocity to a subsequently established magnetic field, and the Onsager-Feynman quantization of superfluid velocity \cite{Onsager, Feynman} are shown to be violated in a two-component superconductor. The manifestation of the two principles normally involves the fundamental constants alone, but this no longer holds as is demonstrated explicitly for the projected liquid metallic states of hydrogen and deuterium at high pressures. The rotational responses of liquid metallic hydrogen or deuterium identify them as a new class of dissipationless states; they also directly point to a particular experimental route for verification of their existence.

Non-classical response to rotation is a hallmark of quantum ordered states such as superconductors and superfluids. The rotational responses of all presently known single-component “super” states of matter (superconductors, superfluids and supersolids) are largely described by two fundamental principles and fall into two categories according to whether the systems are composed of neutral or charged particles. For superfluid systems composed of electrically neutral particles (liquids, vapors, or even solids \cite{Lifshitz}) and for slow rotations, a fraction of the system, the superfluid fraction, remains rotational. However in response to rotation exceeding a certain critical rotation frequency, the superfluid fraction comes into rotation by means of vortex formation. Onsager and Feynman \cite{Onsager, Feynman} pointed out that the superfluid velocity (\(v\)) in these vortices in single-component systems is quantized and the circulation quantum \(K\) depends only on particle’s mass \(m\) and Planck’s constant \(h\): \(K = \frac{1}{2\pi} \oint v \cdot dl = \frac{\hbar}{m}\). Here we mention that for superfluids with e.g. \(p\)-wave symmetry of the order parameter which are also invariant under simultaneous phase and spin transformations this quantization is modified \cite{Babaev}. We also mention that a special situation occur in a multicomponent superfluid with a dissipationless drag (Andreev-Bashkin effect) where a superfluid velocity of one condensate can carry superfluid density of another \cite{Babaev}. Below however we will consider the mixture of charged condensates only with the simplest symmetry of the order parameter coupled by a gauge field.

For systems composed of charged particles and which are also superconducting (electronic Cooper pairs in metals, or protonic Cooper pairs in neutron stars) vortices are not induced by rotation; however, the rotational response of these systems is no less interesting. London showed that a uniformly rotating single component superconductor generates a persistent current in a thin layer near its surface, and this in turn produces a detectable magnetic field, the London field \cite{London}. London related this field to the rotation frequency, \(\Omega\), according to \(\mathbf{B} = -\frac{2mc}{e}\Omega\), where \(m\) is the electron’s mass, \(e\) denotes it electric charge and \(c\) is the speed of light. This law is experimentally confirmed (see e.g. \cite{Kronig} and references therein). Of crucial significance is the fact that the experimentally observed London Law involves only the exact values of the fundamental constants, and not on materials properties specific to the superconductor (such as an effective mass for electrons). This law also holds for electronic superconductors with \(d\)- and \(p\)-wave pairing symmetry.

We here consider the responses to rotation of the projected novel quantum states of metallic hydrogen and metallic deuterium, two-component systems exhibiting off-diagonal long-range order. These are now the subjects of renewed experimental pursuit especially because of the recent breakthrough in artificial diamond technology. The expectation of achieving static pressures in diamond anvil cells perhaps exceeding the expected metallization pressure of hydrogen at low temperatures has now been raised. Liquid metallic states of hydrogen were predicted earlier to exhibit Cooper pairing both in protonic and electronic channels \cite{Kronig}; however it should be noted that an even simpler situation may occur in liquid metallic deuterium because deuterons are bosons and can undergo condensation without the need for a pairing instability. Another possible system where such states may be realized is a hydrogen-rich alloy where under extreme but experimentally accessible pressures both electrons and protons may be mobile in a crystalline lattice \cite{Yakovlev}. Finally a rotational response similar to that discussed below would be present in solid metallic hydrogen or deuterium if it exhibits a metallic equivalent of supersolidity. For brevity below we shall always refer to “liquid metallic hydrogen (LMH)” but it is important to keep in mind that the range of potential applications is much wider, including recent discussions of possible presence of several charged barionic condensates in neutron stars \cite{Egor}. The main motivation of our study is to identify an effect which can provide a possible experimental probe for the renewed experimental search for superconducting...
liquid metallic hydrogen.

It has been observed that because in these systems the charged condensates are replicated twice (e.g. coexistent electronic and protonic, or deuteron, condensates) composite neutral superfluid modes exist [12,13]. These cannot be classified as superconductors in the usual sense; we will see below that also the superfluid mode is quite different from superfluid modes in one-component neutral systems. Previous studies of this state have, however, mostly focused on the reaction of the system to an applied magnetic field [12,14,13]; here our intention is to study the reaction of the system to rotation. The composite superfluid and superconducting modes in this system are inextricably intertwined and as we find below this has unusual manifestations in rotational response, which extend our general understanding of quantum ordered fluids.

The general route to describe a two component superconductor is the London (or hydrodynamic) approach. The system in this approach is described by the following free energy:

\[ F = \frac{1}{2\hbar^2} \sum_{\alpha = e,p} \Psi_\alpha^\dagger (\nabla \theta_\alpha \pm eA)^2 + \frac{1}{2} (\nabla \times A)^2. \]

Here, \( \Psi_\alpha \) and \( m_\alpha \), \( (\alpha = e,p) \) denote electronic and protonic condensate wave functions and corresponding masses and \( A \) is the vector potential. In what follows \( e \) stands for an electric charge of a Cooper pair and we set \( \hbar = 1, e = 1 \). In this work we focus on the effects caused by the coupling to the gauge field and thus we do not consider possible drag effects [6]. Nor do we consider different pairing symmetries.

This model can be rewritten as [12]

\[ F = \frac{1}{2} \left[ \frac{\Psi_e^2}{m_e} + \frac{\Psi_p^2}{m_p} \right] (\nabla (\theta_e + \theta_p))^2 + \frac{1}{2} \left[ \frac{\Psi_e^2}{m_e} + \frac{\Psi_p^2}{m_p} \right] \times \]

\[ \left( \frac{\Psi_e^2}{m_e} \nabla \theta_e - \frac{\Psi_p^2}{m_p} \nabla \theta_p - eA \left[ \left( \frac{\Psi_e^2}{m_e} + \frac{\Psi_p^2}{m_p} \right) \right]^2 + \frac{B^2}{2} \right) \]

The first term here displays no coupling to the gauge field and therefore represents a neutral or superfluid mode which is associated with co-directed flows of electronic and protonic Cooper pairs (with no net charge transfer) [12]. The second term accounts for the superconducting (or charged) sector of the model describing electrical currents. In what follows, we denote a vortex with phase windings \( (\Delta \theta_e = 2\pi n_e, \Delta \theta_p = 2\pi n_p) \) as \( (n_e, n_p) \).

Let us begin with inspection of the composite neutral mode's response to rotation. The simplest topological excitation in the superfluid sector of the model i.e. a simplest vortex which has a nontrivial winding in the phase sum \( (\theta_e + \theta_p) \) is a vortex with the windings of only one of the phases: \( (\pm 1, 0) \) or \( (0, \pm 1) \). We note that since the first term in [1] is symmetric with respect to electronic and protonic condensates, both the \( (1, 0) \) vortex and \( (0, 1) \) vortex have identical configurations of the neutral composite (i.e. consisting of both electrons and protons) superflow. The difference between these two vortices lies only in the contribution to the second term in [1] representing the charged (superconducting) sector of the model.

We first focus on a \( (0, 1) \) vortex. For this case, the solution for vector potential \( A \) at distances from the core much larger than penetration length is given by \[ \frac{1}{2} |A| = \frac{1}{|r|} \frac{\Psi_e^2}{m_p} \left[ \frac{\Psi_e^2}{m_p} + \frac{\Psi_p^2}{m_e} \right]^{-1} \], where \( r \) is the distance from the core center. The superfluid velocities of electrons and protons in such a vortex at a large distance from the core are \( \mathbf{v}_p = (\nabla \theta_p + eA)/m_p \); and \( \mathbf{v}_e = -eA/m_e \). An equilibrium of a rotating system is achieved when the quantity \( E_r = E - \mathbf{M} \cdot \Omega \) is minimal (\( \Omega \) is the rotation frequency and \( \mathbf{M} \) and \( E \) are the angular momentum and energy). Observe that if a system nucleates a vortex \( (1,0) \) then not only protons but also electrons contribute to the angular momentum whose magnitude is given by: \[ |\mathbf{M}| = |\mathbf{M}_e + \mathbf{M}_p| = \int (m_p |\Psi_p|^2 v_p + m_e |\Psi_e|^2 v_e) r dV \]

The superfluid velocity circulations for protons and electrons in a vortex \( (0,1) \) are given by: \[ \oint \mathbf{v}_{e,p} \cdot d\mathbf{l} = 2\pi K(e,p) = 2\pi \left[ \frac{\Psi_{e,p}}{m_{e,p}} \right]^2 \left[ \frac{\Psi_{e,p}^2}{m_{e,p}} + \frac{\Psi_{e,p}^2}{m_{e,p}} \right]^{-1} \frac{1}{m_{e,p}} \]. From this we observe that in the two-component superconductor the Onsager-Feynman quantization rule is violated: the superfluid velocity quantization is fractional and the electronic and protonic circulation quanta \( K_{e,p} \) depend not only on mass but also on densities according to:

\[ K_e = \frac{1}{\frac{\Psi_e^2}{m_p} + \frac{\Psi_p^2}{m_e}} \frac{1}{m_e}; \quad K_p = \frac{1}{\frac{\Psi_e^2}{m_p} + \frac{\Psi_p^2}{m_e}} \frac{1}{m_p}. \]

The quantization conditions [2] holds also for the vortex \( (1,0) \). It has been argued previously that quantization of magnetic flux in LMH is also fractional [12]. The fractionalization of superfluid velocity quantization which we find here has, however, a different pattern. To compare the fractionalization of magnetic flux quantum \( \Phi_0 = 2\pi e \) and the fractionalization of superflow quantization we introduce an angle \( \beta \) as a measure of the ratio of the average condensates densities, as follows: \( \sin^2 (\beta/2) = \left[ \frac{\Psi_e^2}{m_e} + \frac{\Psi_p^2}{m_p} \right]^{-1}; \cos^2 (\beta/2) = \left[ \frac{\Psi_e^2}{m_e} + \frac{\Psi_p^2}{m_p} \right]^{-1} \).

Let \( K_{e,p}^0 = 1/m_{e,p} \) be the standard superflow circulation in a one component neutral superfluid composite of particles with the masses of electronic and protonic Cooper pairs correspondingly. The quantization fractionalization pattern in this notation is then summarized in the Table 1.

The energy per unit length \( E \) of vortices \( (1,0) \) and \( (0,1) \) contains a logarithmically divergent part arising from the
first term in $\mathbf{11 \, 12 \, 13}$:

$$
\mathcal{E} \approx \pi \left[ \sin^4 \left( \frac{\beta}{2} \right) \frac{\Psi_p^2}{m_p} + \cos^4 \left( \frac{\beta}{2} \right) \frac{\Psi_e^2}{m_e} \right] \log \frac{R}{a},
$$

where $a$ is a cut-off length which depends on the core structure and $R$ is the distance from the vortex center to the system boundary. The formation of vortices in response to rotation is controlled by the neutral mode [i.e. by the first term in $\mathbf{11}$]. As discussed above, the vortices $(1, 0)$ and $(0, 1)$ have the same neutral superflow but different contributions to the second term in $\mathbf{11}$. The energetically preferred excitations forming in response to rotation are therefore the $(0, 1)$ vortices which carry a smaller fraction of $\Phi_0$. We remark that composite vortices of the type $(\pm 1, \mp 1)$ do not contribute to superfluid sector of the model and are irrelevant in this rotational physics. On the other hand it is straightforward to show that the vortices $(\pm 1, \pm 1)$ are unstable.

If a vortex $(0, 1)$ is now placed into a cylindrical system with radius $R$ and unit height the system acquires an angular momentum:

$$
|M| = \pi R^2 \frac{|\Psi_e|^2}{m_e} \frac{|\Psi_p|^2}{m_p} \left[ \frac{|\Psi_e|^2}{m_e} + \frac{|\Psi_p|^2}{m_p} \right]^{-1} (m_e + m_p).
$$

Vortices form when $E_r = \mathcal{E} - M \cdot \Omega < 0$. This determines the critical rotation frequency as

$$
\Omega_c \approx \frac{1}{R^2 (m_e + m_p) \log \frac{R}{a}} \log \frac{R}{a} \quad (3)
$$

We can make a rough estimate of critical frequency: $\Omega_c \approx (m_e/m_p)(e^2/a_0)(a_0/R)^2 \log(R/a)$, where $a_0$ is the Bohr radius, which for a $100\mu$m sample is of order of $10\text{Hz}$. Though we deal with a composite superfluid mode and fractional circulation quantization, the critical frequency is approximately the same as it would be in liquid of Cooper pairs of neutral particles with a mass of the proton. However the underlying physics is indeed quite different. One circumstance is that besides fractional quantization of circulation, only a small fraction of the condensates participates in the superfluid mode (its stiffness is $\frac{|\Psi_e|^2}{m_e} \frac{|\Psi_p|^2}{m_p} \left[ \frac{|\Psi_e|^2}{m_e} + \frac{|\Psi_p|^2}{m_p} \right]^{-1}$). Another difference can be seen by considering a similar system but composed of two types of particles with equal masses and charge. This also features a superfluid mode but no vortices can be induced by rotation (this also applies to electronic superconductors where multicomponent order parameters arises from non s-wave pairing symmetry).

A quite deep difference in the rotational physics in two component charged systems is manifested especially in the novel “aggregate states” of vortex matter they should allow. As discussed above, in the simplest case the rotating system forms a lattice of vortices $(0, 1)$ (see Fig. 1A). In this respect the main difference between this system and an ordinary superfluid is that the rotation-induced vortices are also carrying magnetic flux $\Phi = \cos^2(\beta/2)\Phi_0$. The most interesting situation arises when a rotating system is also subjected to a magnetic field. In this case the possible states of vortex matter are numerous and we will consider here some particularly interesting possibilities of novel states of “vortex matter”.

If a weak magnetic field is applied in a direction opposite to the field of rotation-induced vortices the superconducting sector of $\mathbf{11}$ would try to minimize its energy by introducing $(1, -1)$ vortices. These vortices have no neutral superflow (electronic and protonic currents are counter-directed) but carry one magnetic flux quantum $\mathbf{12}$. However a vortex $(1, -1)$ is not stable in a lattice of $(0, 1)$ vortices because it experiences an attraction to such vortices within the range of the penetration length scale $\mathbf{13}$. A vortex $(1, -1)$ therefore should annihilate with a $(0, 1)$ vortex, resulting in a $(1, 0)$ vortex state. At length scales larger than the penetration length a vortex $(1, 0)$ has a Coulomb repulsive interaction with a vortex $(0, 1)$ similar to interaction between two $(0, 1)$ vortices $\mathbf{13}$ and therefore under normal conditions will occupy a space in a rotation-induced lattice of $(0, 1)$ vortices; it can therefore be viewed as a ground state “electronic vortex-impurity” in a “protonic vortex lattice” (see Fig. 1B). The concentration of these “vortex-impurities” depends on the applied magnetic field and there are indeed many interesting possibilities for their orderings and phase transitions.

Consider next a situation with a stronger magnetic field and with a rotation frequency just above $\Omega_c$. Then the dominant structure is a field-induced lattice of composite vortices $(1, -1)$ (as in the case of no rotation $\mathbf{12 \, 14 \, 15}$). Here the energetically most favorable way to introduce a superfluid momentum-carrying vortex is the substitution of one of the $(1, -1)$ vortices by a $(1, 0)$ vortex (see Fig. 1C). This vortex interacts repulsively with its neighbors, carries almost one magnetic flux quantum, but also possesses angular momentum in the superfluid sector. This vortex is therefore an “elementary vortex-impurity” in a lattice of composite vortices. Such a system should exhibit a number of novel phase transitions and vortex matter states. One such transition will occur because there is a finite potential barrier for a “vortex impurity” to jump from one lattice site to another. At certain temperatures the vortex impurities should be able to move from one site to another freely. There is an analogy between “light” vortices, which in the ground state are

| TABLE I: Fractionalization of superflow circulation and magnetic flux quanta. |
|----------------|----------------|
| composite superfluid mode $\sin^2(\beta/2)\Phi_0 - \cos^2(\beta/2)\Phi_0$ |
| electronic superflow circulation $\cos^2(\beta/2)K_c^0 \cos^2(\beta/2)K_p^0$ |
| protontic superflow circulation $\sin^2(\beta/2)K_p^0 \sin^2(\beta/2)K_p^0$ |
be disordered in the vortices the phase of the corresponding condensate will be as an "interstitial vortex defect" (see Fig 1D) which, for a range of parameters, should be a more energetically preferred way to acquire angular momentum than the first possibility. The "light" vortices may form an “interstitial vortex liquid” state, while the co-centricity of light vortices with the lattice of heavy vortices is controlled by a different energy scale. This is again a state with co-existent vortex crystalline order and vortex defect fluidity and yet another example of a “vortex supersolid” which resembles the supersolid state of interstitial particles in crystals discussed in [4].

Finally, let us consider the reaction of the superconducting sector of the system to rotation. It is important to note that electronic and protonic Cooper pair momenta depend on the same vector potential, \( \mathbf{P}_\alpha = \nabla \theta_\alpha = m_\alpha \mathbf{v}_\alpha + e_\alpha \mathbf{A} \) and hence \( \mathbf{A} = \frac{\mathbf{P}_\alpha}{e_\alpha} - \frac{m_\alpha}{e_\alpha} \mathbf{v}_\alpha \) (where \( e_{(e,p)} = \pm e \)). Consider now the situation without an applied external field and low rotation frequencies, so that there are no vortices (i.e. \( \Omega < \Omega_c \)). Then taking the curl of the previous expression we arrive at the constraint dictated by gauge-invariance:

\[
\frac{m_p}{e_p} \nabla \times \mathbf{v}_p = \frac{m_e}{e_e} \nabla \times \mathbf{v}_e. \tag{4}
\]

Let us consider first the zero temperature case when there is no normal component. If the condensate charges entering the problem are opposite (as is indeed the case for LMH) this equation has a trivial solution: \( \mathbf{v}_p = \mathbf{v}_e = 0 \) i.e. at \( T = 0 \) for \( \Omega < \Omega_c \) the condensates remain irrotational. However in the presence of a normal component with a net electric charge its rotation produces an electric current so the superconducting component necessarily has to respond (i.e. \( \mathbf{v}_p = \mathbf{v}_e = 0 \) can no longer be a stationary solution). From (4) it also follows that in contrast to London’s picture for ordinary superconductors [1], superconducting electrons and protons will not follow the rotation of normal component because it would violate constraint (4). This dictates a counter-intuitive situation, namely that in response to slow rotation the superconducting electrons and protons can only move in opposite directions and at different speeds. Their superconducting velocities can be expressed in the following form: \( \mathbf{v}_\alpha = \gamma_\alpha \Omega \times \mathbf{r} \). To find \( \gamma_\alpha \) we first observe that from the stationarity requirement we can obtain an extra condition by equating the rotation-induced electric current of the normal component (multiplied by -1) to the rotation-induced current response of superconducting sector subject to constraint (4):

\[
\mathbf{J}_e = (e_p \gamma_p |\Psi_p|^2 + e_e \gamma_e |\Psi_e|^2) \Omega \times \mathbf{r}.
\]
From the overall electrical neutrality of the system it follows that the rotation-induced normal current is \( \mathbf{J}_n = -(e_p|\Psi_p|^2 + e_e|\Psi_e|^2) \Omega \times \mathbf{r} \). Hence we find

\[
\mathbf{v}_p = \frac{|\Psi_p|^2 - |\Psi_e|^2}{|\Psi_p|^2 + \frac{m_e}{m_p}|\Psi_e|^2} \Omega \times \mathbf{r}; \quad \mathbf{v}_e = \frac{|\Psi_e|^2 - |\Psi_p|^2}{|\Psi_e|^2 + \frac{m_p}{m_e}|\Psi_p|^2} \Omega \times \mathbf{r}.
\]

To sustain these counter currents a rotating two-gap superconductor should generate in its bulk a vector potential and hence rotation induces a magnetic field:

\[
\mathbf{B}_{rot} = \frac{2}{e} \frac{(|\Psi_p|^2 - |\Psi_e|^2)}{m_p|\Psi_p|^2 + |\Psi_e|^2} \Omega.
\]

While in the bulk the superfluid electrons and protons have the velocities \( \mathbf{v}_{e,p} \), the field \( \mathbf{B}_{rot} \) is generated by velocity variations in the layer near the system’s edge with the thickness of the penetration length \( \lambda = (e^2/|\Psi_p|^2/m_p + |\Psi_e|^2/m_e)^{-1/2} \). This follows from the equation for magnetic field variation, namely:

\[-\nabla^2 \mathbf{B}(\mathbf{r}) + \mathbf{B}(\mathbf{r}) = \mathbf{B}_{rot}. \]

Eq. (5) demonstrates a remarkable circumstance: the London Law in the two-component superconductor is actually violated. The rotation-induced field is not a universal function of the fundamental constants irrespective of microscopic details. Indeed it acquires a dependence on densities. At temperatures just below superconducting transition for protons a rotating sample of radius \( R \) generates a magnetic flux of order of \( R^2 \Omega \) flux quanta (\( R \) given in cm and \( \Omega \) in s\(^{-1}\)) which could be detectable with modern SQUIDs even for samples as small as 10\( \mu \) rotating at 1Hz (we note that it is easier to achieve high pressures in small samples which makes it a very convenient experimental probe). Going to a larger sample or higher rotation frequency would even allow measurement of rathio of the condensates densities and their temperature dependences, as follows from [5]. And, of course, its absence would even rule out protonic superconductivity or deuteron condensation. It follows that a direct experimental route exists for the verification of this possible new class of dissipationless states.

Though this has been cast in terms of a possible failure of London’s law (otherwise rigorously applicable up to relativistic corrections in electronic superconductors) the major issue discussed here might well be viewed as a possible extension of the classifications of the rotational responses of quantum fluids. Rotational response is a quintessentially state-defining property of quantum fluids, and the one we find in LMH (as summarized in Figure 2) is seen to be quite complex, and it involves both co- and counter-directed electrical currents, and in particular a current in the direction opposite to rotation. This suggests a classification of the projected liquid state of metallic hydrogen as a new quantum fluid, and one which may be presenting considerable opportunity for new and emerging physics. From an experimental point of view it now appears that the rotational response of multicomponent superconductors may well offer the most direct probe, both qualitative and quantitative, of the corresponding quantum orderings of hydrogen and deuterium in experiments in diamond anvil cells. It may also be extendable to the ternary systems formed by addition defects in crystals.

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