Observation of a partially rotating superfluid of exciton-polariton

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Rotation of a container holding a viscous fluid forms a vortex which grows with increasing angular velocity. A superfluid, however, is intrinsically different from these normal fluids because its rotation is quantized. Even if a container of superfluid is rotating, the fluid itself remains still until a critical velocity is reached. Beyond the critical velocity, all the particles conspire to suddenly pick up an angular momentum of \( \hbar \) each and forms a quantized vortex. As a result, a superfluid is known to increase its rotation by a total angular momentum of \( N\hbar \) only. In this letter, we show that exciton-polariton superfluid can split into an irrotational part and a rotational part. The relative ratio between the two states can be controlled by either pump beam’s power or spot size. Consequently, angular momentum of exciton-polariton superfluid can be tuned from zero to \( N\hbar \) continuously. This striking observation sets the stage for studying non-equilibrium properties of a superfluid with exciton-polaritons.

A superfluid is typically referred to as a manifestation of macroscopic quantum coherence. In other words, a superfluid can be described with a macroscopic and coherent single wave function, \( \psi(r,t) = \psi_0(r,t)e^{i\phi(r,t)} \), where the amplitude \( \psi_0 \) and phase \( \phi \) of the wave function are both given as functions of position \( r \) and time \( t \). An important aspect of this is the formation of quantized vortices.

If one assigns a single wave function, \( \psi(r,t) \), to the superfluid state, the superflow velocity, \( v \), turns out to be proportional to the phase gradient of the wave function, \( v = \nabla \phi(r,t) \). Since the phase factor is a scalar field, the curl of superflow velocity is zero, that is, superfluid is irrotational.

One way to make a superfluid rotate is to create a vortex whose core is void of superfluid. Around this core, circular phase gradient can be produced and the fluid can thus flow in circle. Since the wave function must be single valued, the phase difference along a closed loop around the vortex core must be an integer multiple of \( 2\pi \) and therefore the speed of circulating superfluid is given by \( n\hbar/mr \). Here \( \hbar \) is the reduced Planck constant, \( m \) is the mass of the particle, \( r \) is the radius of a circular flow, and \( n \) is an integer number indicating the amount of quantized flow. On that account, each particle in the superfluid state is known to carry identical angular momentum \( n\hbar \). Because all the particles take the same angular momentum value, the total angular momentum of the fluid quantized by integer multiples of \( N\hbar \), where \( N \) is the number of particles.

These vortex states are ones of the excited states within which the single wave function description and thus superfluidity is preserved. In other words, vortex states are not a result of viscous excitations that are typically described with two fluid model of superfluid. Therefore, the vortex states have been viewed as the states where the entire superfluid gets excited into simultaneously. For example, an angular acceleration of a container filled with superfluid goes through an abrupt transition from a zero angular momentum state to a finite angular momentum state of \( N\hbar \) at the critical velocity.

However, we found that it is possible to rotate a fraction of a superfluid in an exciton-polariton system. This happens by the superfluid splitting into different states carrying different amount of angular momentum. Individual particle’s angular momentum is still quantized by \( \hbar \), but the total angular momentum of the system as a whole is no longer quantized by \( N\hbar \), in contrast to what was previously observed in more conventional single component bosonic superfluids such as liquid helium or cold atomic gases.

The experiment is carried out following a fairly standard non-resonant pumping of a GaAs quantum well microcavity with a Laguerre-Gaussian (LG) beam as depicted in Fig. 1. A pair of distributed Bragg reflector (DBR) forms an optical cavity of \( \lambda/2 \) with a set of GaAs/AlAs quantum well stacks. The set consists of four stacks of quantum wells and is situated at the antinode of the cavity. At the positions of the two antinodes on either side of the cavity, identical sets of quantum wells are also placed to increase overlap between the excitons and electric fields. The sample is exactly the same as that used in the reference. The sample is placed inside a cryostat with an optical window reaching as low as 6 K.

A mode-locked Ti: Sapphire pulse laser with 1.72 eV (720 nm), a considerably higher energy than the QW, is used to non-resonantly pump electron-hole plasma into the QWs. The width and repetition rate of the pulse is 150 fs and 80 MHz, respectively. The LG beam is generated by pushing a Gaussian laser beam through a 2\( \pi \) winding phase-
mask. When the LG beam is focused onto the sample, the orbital angular momentum of the beam is transferred to the sample.

Figure 1. Experimental setup and two different states of polariton. a, Schematic image illustrating our findings reported in this letter. A sample is excited by non-resonant LG beam laser of 1.72 eV. Above threshold pumping power, polariton condensates are formed into two different energy states with different spatial distribution. b, Schematic of the experimental setup. LG beam laser, which generated by optical phase mask, excites the sample in a cryostat (6 K). Photoluminescence (PL) is measured by monochromator and CCD camera with Mach-Zehnder interferometer. c, d, Energy-momentum dispersion of polaritons below and above the threshold pumping power respectively. e, Energy-integrated PL image of polaritons above the threshold pumping power. f, Energy-space dispersion for a given slice of polariton emission, marked by white dotted lines in e by blocking one arm of Mach-Zehnder interferometer. g, Interference images are obtained by setting up a modified Mach-Zehnder interferometer in the same situation as e. A $2\pi$ phase winding is evidenced by the fork pattern shown in g. h, Energy-space dispersion with the interference pattern.

This method is known to create quantized vortices in exciton-polariton condensate which is evidenced by a forking of the interference pattern at the vortex core. A standard interference technique is used to generate energy integrated emission in real space. The fact that $2\pi$ phase-winding is seen in the interference pattern in Fig. 1g implies that at least some part of this superfluid carries an angular momentum of $\hbar$ per particle. However, energy dispersion shown in Fig. 1f reveals that the condensate consists of multiple energy states. The lowest energy state observed in our experiment is formed near $k_y = 0$ indicating that the lowest energy state is indeed the ground state condensate and not the vortex carrying state.
In order to investigate this phenomenon in more detail, we employed spectroscopic interferometry or energy-resolved spatial interferometry (ERSI) based on a fairly standard tomographic measurement protocol. The experimental procedure is almost identical to the modified Mach-Zehnder interferometry commonly used to image quantized vortices. One big difference in this setup is that the interference pattern is passed through a narrow slit and the selected slice is then injected to a monochromator. We obtain interference pattern in the energy-position space as shown in Fig. 1h. By blocking one arm of the interferometry set-up one can easily obtain the energy-integrated PL image and the energy-position diagram for emission intensity without an interference pattern (see Fig. 1e and Fig. 1f, respectively). The energy-position diagram clearly displays more than one distinct energy states, which is well consistent with the dispersion spectrum shown in Fig. 1d.

The system being distributed over multiple energy levels were observed in other experiments before what is unique about the excited state seen in our experiment is that it has to be a vortex carrying state since the ground state, by definition, cannot carry any vortex. To experimentally verify this, we constructed the full ERSI pattern by scanning the slit through the original interference image and combining all of these slices together. Once the ERSI pattern is obtained, one can look at a cut of real space interference for a given value of energy. Since the energy levels are quantized in this spectrum, we look at each of these quantized energy levels separately (See Fig. 2a,d). Fig. 2b, e shows the interference pattern with $E = 1592.37$ meV and $1591.69$ meV.

![Fig. 2. Energy-resolved PL image, interference, phase of two states. By combining tomographic measurement and interferometry, energy-resolved PL image and energy-resolved interference can be obtained.](image)

Interference patterns from the two different energy levels are distinctly different as shown in Figs. 2b and 2e. The ground state shows simple stripes of a phase coherent light as expected (Fig. 2e). In contrast, the excited state shows a dislocation line in the interference pattern indicating a singularity and a $2\pi$ phase winding about that
singularity (Fig. 2b). This shows unambiguously that the superfluid somehow is split into two components with two different angular momenta, the ground state having zero angular momentum and the excited state having $\hbar$ per particle.

We can deduce relative population between the vortex carrying state and the ground state from the intensity measurement of Fig. 3a. It shows the energy spectrum as a function of the radial position with color code indicating the intensity of the emission. Integrated intensity of each energy state is proportional to the ground state population, $n_g$, and the excited state population, $n_e$ (Fig. 3b). The fraction of the irrotational superfluid, $r_L$, and rotating superfluid, $r_e$, can be easily calculated according to $r_{g,e} = \frac{n_{g,e}}{n_g + n_e}$ (Fig. 3c).

Coexistence of vortex carrying excited state and the stationary ground state may be related to non-equilibrium nature of exciton-polariton condensates. Even though incident laser transfers its angular momentum to the sample, the equilibrium state of the non-resonantly pumped exciton-polariton condensate is probably that of the vortex non-carrying ground state as the incoherent scattering processes during the relaxation would carry away all the energy, linear and angular momentum of the original particles. The vortex carrying state that are being observed is most likely an intermediate state whose angular momentum has not fully relaxed to zero yet.

The fraction of rotating superfluid can be controlled in two ways. The first method is to change the power of the pump beam. As one increases the pump beam’s power, the fraction of ground state superfluid reaches about 50% at its maximum near the threshold pumping power. However, with increasing the power further, the ground state portion is reduced down to about 12%. Similar effect was seen with vortices created by chiral lens.

The fact that fraction of each state is not a constant of pumping power indicates that non-linear decay process from the excited state into the ground state is involved. Otherwise, a constant decay rate independent of pumping power would hold the fraction of each state a constant. A stimulated scattering into the vortex-carrying state could enhance the fraction of the state at a higher pumping power since more particles are present in the excited states. With increasing the pumping power there is a massive blueshift and level broadening due to the repulsive interaction between exciton reservoir and polaritons. It is conceivable that this repulsion and the stimulated scattering is balanced out at about five times the threshold power and the relative population levels beyond this pumping power (See Fig. 3c).

Figure 3. Energy-space dispersion under different pumping power. a, From the left, each data show energy-
space dispersion when 0.72, 1.0, 1.8, 2.8, and 10 times the threshold power (1.8 mW). Below the threshold, a continuous energy and spatial distribution is seen. Above the threshold, the polariton condenses into two states, which are maintained beyond the 10 times the threshold. The blueshift of polaritons is observed with increasing pump power due to the repulsive interactions between exciton reservoir and polaritons. b, Populations of the ground state (red circle) and excited state (blue square) polaritons are plotted against the pumping power. The population is obtained by integrating the PL intensity in each states. c, Relative population between the ground and excited state. At the threshold, more polaritons are found in the excited state. As the pump power increases slightly above the threshold, the fraction of the excited state population decreases rapidly and the population of the ground state and excited state reaches to roughly around 50-50 at \( 1.2 \frac{P}{S} \). Beyond \( 1.2 \frac{P}{S} \), the fraction of excited state polaritons increases reaching as high as 80%.

Another method for increasing the amount of rotation is to control the pump beam’s spot size as shown in Fig. 4. The relative population is plotted against the size of the pumping beam in Fig. 4b. For a relatively small pump beam diameter of 5.8 \( \mu \text{m} \), less than 30% of the polaritons are found in the vortex carrying state. As the LG beam size increases, the relative population of the vortex carrying state increases fairly rapidly reaching about 85% at about 7.1 \( \mu \text{m} \) diameter. We note that even higher energy states are excited for larger ring size.

For the ground state condensate to form near the center of the LG beam, sufficient amount of polaritons must flow in so that the critical density for condensation is reached. Once the ground state condensate is formed, stimulated scattering, this time towards the ground state, may speed up the relaxation. This process would be more favorable for smaller ring size pump beams which is exactly what is observed. Consequently, larger amount of blueshift is observed for smaller beam size with increasing density near the center (Fig. 4c).

Figure 4. Energy-space dispersion under different pumping laser diameter. a, From the left, each data shows energy-space dispersion for the diameters of 4.8, 5.8, 6.3, 7.1, and 8.5 \( \mu \text{m} \). The solid white lines are the intensity profile of the pumping LG beam, and pumping laser diameter is identified with the distance between two maximum points of the laser profile. When the diameter is small, the ground state dominates, but as the LG beam size increases, the excited state population increases more rapidly. When the LG beam size becomes very large, the ground state almost disappears, and even 2nd excited state starts to appear. b, Portion of polaritons in each state with respect to the LG beam diameter. The ground state (red square) is dominant at small diameters, but decreases rapidly as the LG beam size increases. And completely suppressed above about 7.1 \( \mu \text{m} \). The excited state become dominant as increasing diameter. However, the excited state does not consist of a single level with large LG beam diameters. Above 7.1 \( \mu \text{m} \), the excited state splits in to the first excited state (blue circle) and the second excited state (green diamond). The relative population between these excited states is also affected by laser beam diameter. c, Energy of each state is plotted against the pumping laser diameter. Energy of all the states tend to decrease as
the LG beam size increases due to the lower polariton density and thus smaller amount of blueshift. Energy of the excited state at larger laser diameters can be less than the energy of the ground state at smaller laser beam sizes.

In conclusion, exciton-polariton can be partially rotated in its superfluid state and the relative portion of the rotating component can be experimentally controlled. The total angular momentum of the superfluid can therefore be tuned continuously as well. This is the first time such effect is seen in a single component bosonic superfluid. The striking result likely owes its origin to exciton-polariton’s fundamentally non-equilibrium nature where we are observing the time averaged image of the transient relaxation process between the vortex-carrying state and the ground state. It would be interesting to carry out time resolved measurement of the energy relaxation in order to reveal this transient nature and explore the non-equilibrium properties of the exciton-polariton superfluid in general.

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Author contributions
D.C. and M.P. carried out the experiments and data analysis. D.C. and M.P. contributed equally to this work. B.Y.O. and M.S.K. contributed to the experiment at an early stage. The sample was grown by S.I.P., S.K., and J.D.S.. Y.H.C. and H.C. conceived the project and supervised its development. All authors have discussed the results and contributed to editing this manuscript.

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