Controlling free superflow, dark matter and luminescence rings of excitons in quantum well structures

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Following the discovery of Bose-Einstein condensation (BEC) in ultra cold atoms [E. Gosta, Nobel Lectures in Physics (2001-2005), World Scientific (2008)], there has been a huge experimental and theoretical push to try and illuminate a superfluid state of Wannier-Mott excitons. Excitons in quantum wells, generated by a laser pulse, typically diffuse only a few micrometers from the spot they are created. However, Butov et al. [1] and Snoke et al. [2] reported luminescence from indirect and direct excitons hundreds of micrometers away from the laser excitation spot in double and single quantum well (QW) structures at low temperatures. This luminescence appears as a ring around the laser spot with the dark region between the spot and the ring. Developing the theory of a free superflow of Bose-liquids we show that the macroscopic luminescence rings and the dark state observed in [1, 2] are signatures of the coherent superflow of condensed excitons at temperatures below their Berezinskii-Kosterlitz-Thouless (BKT) transition temperature [3]. To further verify the dark excitonic superflow we propose several keystone experiments, including interference of superflows from two laser spots, vortex formation, scanning of moving dipole moments, and a giant increase of the luminescence distance by applying one-dimensional confinement potential. These experiments combined with our theory will open a new avenue for creating and controlling superflow of coherent excitons on nanoscale.

The dramatic appearance of luminescence rings with radii of several hundred microns in quantum well structures has been originally attributed to a boundary between a positive hole gas diffusing from the laser spot and a negative electron gas located well outside the spot [3]. This implies an energetically unfavorable separation of the charges into hole-dominant positive and electron-rich negative regions on a macroscopic scale. The charge separation model reproduced some basic features of the luminescence ring formation. However, there are several observations that are not accounted for by this simple model including a non-monotonic dependence of the ring radius on laser power and significant short-range Coulomb interactions of the carriers [4]. The model is also refutable on the grounds of a rather short exciton formation time, typically ten picoseconds or less for relevant densities (> 10^{10} cm^{-2}) of photo-carriers [5, 6]. Indeed, photoelectrons (mass m) almost immediately emit optical phonons with the frequency ω, so that their speed is capped at about v = 2bf_{\omega}/m \approx 4 \times 10^{7} cm/s. Thus, the exciton formation mean-free path turns out to be only a few microns, which is too short for creating the charge separation on the macroscopic scale of a few hundreds of micrometers far outside the spot. This is consistent with measurements of a photoluminescence (PL) spectrum in the magnetic field which have shown a diamagnetic shift of the PL energy peak, indicating that the electrons and holes are bound into thermalized pairs (i.e excitons) in the region about a few tens of micrometers from the center of the spot [10]. As one gets closer to the center of the spot, this shift is changed from blue to a red one indicating an electron-hole plasma inside the spot, where electron-hole pairs rapidly form hot excitons by emission of phonons. Therefore, excitons, which can condense in a coherent superfluid, are formed from electron-hole plasma at rather short distances from the center of the laser spot, Fig. (1).

Importantly, since the electron-hole plasma expands inside the spot due to the Coulomb repulsion [11], energy and momentum conservation requires that excitons initially form in states with finite center-of-mass momenta, K, which do not couple to light directly. If the lattice temperature, T is below the Berezinskii-Kosterlitz-Thouless (BKT) transition temperature, T < T_{K_{BKT}} = \hbar^2n_{ex}/k_{B}m_{ex}, which is about a few Kelvin for the relevant exciton densities, n_{ex} \geq 10^{10} cm^{-2}, part of excitons should form a coherent state with a certain value of their momentum, K = K_0 taken from electrons and holes forming pairs. Thus one can expect transitions of electrons and holes from their plasma into a flowing superfluid state of composed bosons in contrast to the more conventional stationary BEC of ultra cold atoms. As a result we expect an excitonic radial supercurrent appearing at some distance, R_0 inside or just around the laser spot, J_s(R_0) \equiv J_0 \neq 0. This new state of matter allows excitons to travel on a macroscopic distance from the laser spot producing PL far away from the photo-excited region (Fig. 1), as observed in Refs. [1, 2].

Model

To describe the concomitant free superflow of two-dimensional (2D) Bose-liquid of excitons we apply a generalized Gross-Pitaevskii equation [13] for the order-parameter, ψ(R) taking into account the exciton recom-
where $\phi$ decay rate, Eq. (3) is the continuity equation with the dimensionless Landau equation in the presence of the current and $\beta = (\xi r)_{\text{diss}}^{\text{exc}}$ coordinates equations $K_\text{as soon as their momentum drops below } K_c$ and they emit photons.

combination rate $\gamma/\hbar$, as originally proposed by Keldysh [14]:

$$ - \left( \frac{\hbar^2}{2m_{\text{exc}}} \Delta + \mu \right) \psi(R) + V|\psi(R)|^2 \psi(R) - i \frac{\gamma}{2} \psi(R) = 0. $$

Here $\mu$ and $V$ conveniently parameterize the average superfluid density and the short-range repulsion, respectively.

Introducing dimensionless amplitude, $f(r) = (V/\mu)^{1/2}|\psi(R)|$, and the current density, $j(r) = f(r)^2 \nabla \phi(R)$, Eq. (1), is reduced to the following two equations

$$ - \Delta f - f^3 + \frac{j^2}{f^3} = 0, $$

and

$$ \nabla \cdot j = -\beta f^2. $$

where $\phi(R)$ is the phase of the order parameter and coordinates $r = R/\xi$ are normalized on the coherence length $\xi = (\hbar^2/2m_{\text{exc}}\mu)^{1/2}$. Eq. (2) is the familiar Ginzburg-Landau equation in the presence of the current and Eq. (3) is the continuity equation with the dimensionless decay rate, $\beta = \gamma/2\mu$.

These equations are grossly simplified in the case of a low decay rate, $\beta \ll 1$. In this case the amplitude of the order parameter and the current density change at the length scale of the order of $1/\beta \gg 1$ much larger than the coherence length, so that the first term ("kinetic pressure") in Eq. (2) is negligible. Then the current density is expressed via the amplitude as

$$ j \approx f^2 \sqrt{1 - f^2}. $$

There are two branches described with Eq. (4), but only one, which counter-intuitively corresponds to a larger order parameter $f^2 \geq 2/3$ and smaller momentum $k = \sqrt{1 - f^2} \leq 1/\sqrt{3}$, is stable for small enough $\beta$, Fig. 2 (see supplementary material).

A. One-dimensional (1D) superflow and PL.

Let us first discuss the stable superflow in 1D structures which can be made by applying a 1D confinement potential to experimentally available 2D samples. By using the boundary condition, $j(0) = V(m_{\text{exc}}/2\mu)^{1/2} J_0$, and assuming that $R_0/\xi \ll 1/\beta$, Eq. (3), written in the form $dk/dx = \beta(k^2 - 1)/(1 - 3k^2)$ for the exciton momentum $k(x)$, can be easily analytically solved:

$$ 3(k_0 - k) + \ln \left[ \frac{(1 + k)(1 - k_0)}{(1 - k)(1 + k_0)} \right] = \beta x, $$

where $k_0$ is found from $k_0 - k_0^3 = j(0)$.

To determine location of luminescence, we use that excitons can radiate in 1D and 2D by resonant emission of photons only if their momentum is inside the photon cone [13, 16], namely if superfluid center-of-mass momentum
is small enough: $K \lesssim K_e = E_g \sqrt{\epsilon/c}$. Here $E_g$ is the semiconductor band gap and $\epsilon$ is the dielectric constant. The exciton PL intensity is determined by the fraction of excitons inside the cone, where their decay rate $\beta$ is strongly enhanced [13]. For a narrow photon cone, $K_e \ll K_0$, the position of a bright luminescence stripe, $x_{\text{ring}}$, is found by taking $k = 0$ in Eq. (5),

$$x_{\text{ring}} = \beta^{-1} \left[ 3k_0 - \ln \left( \frac{1 + k_0}{1 - k_0} \right) \right]. \quad (6)$$

Since $k_0$ is of the order of one, the distance $x_{\text{ring}}$ is macroscopic, i.e. large compared with the coherence length, as $1/\beta \gg 1$.

**B. Controlling PL in 2D superflow.**

Let us now consider an already implemented experimental situation [1, 2], where excitons created around the central laser spot propagate freely in a plane. In contrast to the original proposal of Ref. [1], we suggest a continuous superflow of excitons out of the excitation spot under steady-state photoexcitation, rather than their normal drift and diffusion. For isotropic 2D superflow the order parameter depends only on the distance $\rho$ from the spot. Then 2D continuity equation (3) (written for the exciton momentum $k(\rho) = \sqrt{1 - f^2(\rho)}$) takes a simple one-dimensional form,

$$\frac{dk}{d\rho} + \frac{k - k^3}{\rho (1 - 3k^2)} = \beta \frac{k^2 - 1}{1 - 3k^2}. \quad (7)$$

The stable numerical solution of this equation is shown in Fig. 3 together with $k(x)$ dependence of the momentum in 1D structures using Eq. (5). The analytical solution of linearized equation (7), $k_0' + k/\rho = -\beta$, which is $k(\rho) = k_0 \rho (\rho - \beta \rho)^2$ practically coincides with the numerical solution of the nonlinear Eq. (7) in the whole relevant space ($\rho_0$ is the distance from the center of the spot where the superflow starts). As in the 1D case, PL ring radius corresponds to $k(\rho_{\text{ring}}) = 0$, so that $\rho_{\text{ring}} = (2k_0 \rho_0 / \beta)^{1/2}$. However, different from 1D geometry, where $x_{\text{ring}}$ scales as $1/\beta$, the ring radius scales as $\rho_{\text{ring}} \propto 1/\sqrt{\beta}$ in the 2D case (see inset in Fig. 3). Therefore, the radius of PL for 2D case occurs to be much shorter than the distance between the laser spot and 1D luminescence stripe. This prediction can be verified by channeling the superfluid, that is by applying 1D confinement potential in order to controllably transform 2D to 1D geometry and check if PL shifts away from the spot.

In dimensional units the PL ring radius is $R_{\text{ring}} \approx \sqrt{2h \tau_0 K_0 / (m_{\text{ex}} \alpha)}$. The distance from the center of the spot, $R_0$, where the superflow starts with the momentum $K_0$, can be estimated using the energy conservation. The average electron potential energy inside the spot is $V_n$, where $V \approx 2\pi \alpha d$, $d$ is the effective separation between the electron and hole layers ($\approx 10$ nm), $\alpha = e^2/(4\pi \epsilon_0 \epsilon) \approx 2.3 \times 10^{-28}Jm$, and $\epsilon$ is the static dielectric constant of the semiconductor. It transforms into the exciton kinetic energy, i.e. $V_n = K^2/2m_{\text{ex}}$, so that $K_0 \approx \sqrt{4\pi \alpha d n m_{\text{ex}}}$. The radius $R_0$, where the superfluid is formed is about $R_0 = \sqrt{\gamma^2 \tau_{\text{ex}}}$, where $\tau_{\text{ex}}$ is the exciton formation time. As a result we find

$$R_{\text{ring}} \approx \frac{\hbar \nu_{\text{ex}} (16\pi \alpha d n m_{\text{ex}})^{1/2}}{\gamma} \quad (8)$$

Importantly, $\gamma$ in this estimate is the nonradiative recombination rate since the coherent excitons have their momentum outside the photon cone. The nonradiative lifetime, $\tau_0 = \hbar / \gamma$, is an order of magnitude or more longer than the exciton radiative lifetime [17]. Remarkably, using the realistic material parameters $\tau_{\text{ex}} = 1$ ps, $\tau_0 = 10$ $\mu$s [17], $m_{\text{ex}} = 0.2 m_e$, $\epsilon = 13$ in Eq. (8) yields $R_{\text{ring}}$ about 500 $\mu$m for the photoelectron density $n = 10^{10}$ cm$^{-2}$, explaining the macroscopic radius of the rings. Dissipation processes stabilize the steady-state superflow of coherent excitons (see supplementary material), in contrast with a normal state flow, where individual excitons can scatter to a lower energy state emitting acoustic phonons.

Also the dependence of the ring size and its PL inten-


sity, $I$, on the excitation power, $P$, can be readily understood in the framework of the free superflow. According to Eq. (3) the ring radius scales as $R_{\text{ring}} \propto n^{1/4} \tau_{\text{ex}}^{1/2}$. The ring luminescence intensity, $I$, is proportional to the exciton density on the ring, $n_{\text{ex}}$. Excitons are pumped into the coherent state at $R_0$ with the rate $n/\tau_{\text{ex}}$, where the exciton formation time, $\tau_{\text{ex}}$, is inverse proportional to the photocarrier density $n$ and it strongly depends on the exciton momentum $K_0$ [6]. After flowing the distance $R_{\text{ring}}$ excitons decaying radiating light, so their density on the ring scales with the photocarrier density as $n_{\text{ex}} \propto R_0 n / R_{\text{ring}} \tau_{\text{ex}}$. Parameterizing the momentum dependence of the exciton formation time as $\tau_{\text{ex}} \propto K_0^2 / n$ yields the following scaling: $R_{\text{ring}} \propto n^{(r-1)/4}$ and $I \propto n^{(5-r)/4}$. Hence, we find $R_{\text{ring}} \propto P^{(r-1)/4}$, $I \propto P^{(5-r)/4}$ in the case of a linear photoelectron population ($n \propto P$), and $R_{\text{ring}} \propto P^{(r-1)/8}$, $I \propto P^{(5-r)/8}$ in the case of $n \propto P^{1/2}$. Since $r$ can be a large number ($\geq 5$) [6], our theory predicts an increase of the ring radius and a decrease of the PL intensity with higher excitation power. In particular, the product $R_{\text{ring}} I$ is proportional to $n$ for any exponent $r$ independent of modeling the exciton-formation-time, thus resulting in $R_{\text{ring}} I \propto P^{1/2}$ as observed [1] for $n \propto P^{1/2}$. More generally the dependence of $\tau_{\text{ex}}$ on the exciton momentum is essentially non-monotonic [6], so that the ring radius and the PL intensity may depend on the excitation power in a more complicated fashion as also observed [6].

There is a threshold density of photoelectrons $n_c$ in our theory in agreement with the experiments [1,2], and hence the laser power below which the rings do not form, which can be found from $K_0 = K_c$. At this and lower excitation power coherent exciton superfluid radiates just at the point of its formation inside or close to the laser spot. Using our estimate of $K_0$ one obtains $n_c = E_0^2 / (4 \pi c^2 \alpha m_{\text{ex}}) \approx 0.2 \times 10^{10}$ cm$^{-2}$.

While a detailed description of the outer ring fragmentation into a periodic array observed in Ref. [1] is outside the scope of the current paper, the periodic patterns are anticipated in our theory since the uniform coherent state of the outer ring is unstable due to a strongly enhanced recombination rate inside the photon cone (see supplementary material).

C. Two-spot superflow and dark matter.

Experiments with two rings created by spatially separated laser spots revealed that the rings attract one another, deform, and then open towards each other [3]. This happens before the rings coalesce into a common oval-shaped ring, suggesting the existence of a “dark matter” outside the rings that mediates the interaction. Such a ring attraction is hard to consistently interpret in the classical electron-hole plasma model [11], where outer electron-rich regions from two spots naturally should repel each other. Here we show that this dark matter is the coherent superflow of excitons from two laser spots interfering with each other. Our continuity equation [6] becomes now an equation for the two components of the exciton momentum $k = (k_x, k_y)$,

$$\frac{\partial(1-k^2)k_x}{\partial x} + \frac{\partial(1-k^2)k_y}{\partial y} = \beta(k^2 - 1),$$

(9)

Using $k = \nabla \phi$ it can be reduced to a nonlinear partial differential equation for the phase of the order parameter $\phi(x, y)$ with the boundary condition $k = k_0$ on two small circles inside or just around every spot. As numerically shown above, the linearized continuity equation describes well the superflow from a single spot on the relevant distance, Fig. (3), so that we consider a linearized version of Eq. (9):

$$\Delta_2 \phi(x, y) = -\beta,$$

(10)

where $\Delta_2 = \partial^2 / \partial x^2 + \partial^2 / \partial y^2$. The individual ring size is about $\sqrt{k_0 \rho_0 / \beta}$, so that one expects two independent PL rings when the distance $d$ between the spots is large $d \gg \sqrt{k_0 \rho_0 / \beta}$. The interaction should appear at the distance comparable to the ring size. If we assume that the superflow starts at a small distance from the centers of the spots such that $\rho_0 \ll k_0 / \beta$, then a solution of Eq. (10)

$$\phi(x, y) = -\beta x^2 + y^2 + k_0 \rho_0 \ln(\rho_1 \rho_2)$$

(11)

approximately satisfies the boundary conditions for $d/2 \lesssim \sqrt{k_0 \rho_0 / \beta}$. Here $\rho_{1,2} = \sqrt{(x \pm d/2)^2 + y^2}$ is the distance from each spot, respectively. If we further assume that $\rho_0 \lesssim k_0^2 / (\beta k_0)$, then the condition for PL, $k \ll k_c$, is satisfied at the contour found from

$$\frac{1}{(x - d/2)^2 + y^2} + \frac{1}{(x + d/2)^2 + y^2} = \frac{\beta}{2k_0 \rho_0},$$

(12)

and shown in Fig. 4. The series of the obtained contours agree well with the experimental observations [3]: as the spots are brought closer, the rings deform, and then open towards each other. This happens well before the rings coalesce into a common oval-shaped ring (right lower panel in Fig. 4).

Apart from the normal state diffusion in the charge-separation model, an alternative “superfluid” interpretation of PL rings in double and single QWs has been hampered by the lack of the theory of the free 2D superflow. Based on our theory, we suggest that the macroscopic luminescence rings and the dark matter in QWs originate in the slowly decaying superflow of excitons out of the laser spot at temperatures below their BKT temperature. The existence of a critical temperature about a few Kelvin of the macroscopic ring onset [5] strongly supports our conclusion. Our results imply that Butov et al. [1] and Snoke et al. [2] discovered a new state of matter: the exciton superflow in the dark state. This state can be further experimentally verified with light scattering, STM or any other spectroscopy sensitive to the moving
FIG. 4: (Color online) PL contours from two laser spots for different distances $d$ between the spots (measured in $\xi/k_0 \rho_0/\beta$) obtained by using Eq. (12). As in the experiment [5] the rings first attract one another and, then, open towards each other. See also animations, where PL patterns from two laser spots separated by a fixed distance evolve with increasing $k_0 \rho_0/\beta$ from 0.01 to 0.18 [18].

Dipole moments. Another piece of evidence for the superflow should be an observation of a quantum interference and/or vortex structures with the characteristic coherence length $\xi = \hbar/(2m_e n_e V)^{-1/2}$ of about 15 nm. Last but not least prediction of the theory is a giant increase of the distance to the luminescence region by applying a 1D confinement potential tuning the geometry from 2D to 1D.

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Supplementary material: instability analysis

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Instabilities of the stationary superflow can be analyzed using a generalised time-dependent Gross-Pitaevskii equation, written in our units as

\[-(\Delta + 1)\psi(r, t) + \psi^2(r, t) + \psi(r, t) - i\beta \psi(r, t) = (i - u) \frac{\partial \psi(r, t)}{\partial t}\]

Here \(u\) accounts for any kind of dissipation, for instance emission of phonons as in the time-dependent Ginzburg-Landau equation \([1]\).

One can linearize this equation with respect to small fluctuations of the uniform superflow \(\psi_s(r) = f(r) \exp[i\phi(r)]\) satisfying the stationary GP equation. Taking \(\psi(x, t) = \psi_s(x) + \tilde{\psi}(x, t)\) and \(\dot{\psi}(x, t) = \eta(x, t)\psi_s(x) + \eta'\) yields for small \(|\eta(x, t)|\ll 1\)

\[-\eta''_{xx} - 2\eta'_x (f^{-1} f'_x + i\phi'_x) + f^2 (\eta + \eta^*) = (i - u) \eta \quad (2)\]

in 1D geometry. The stationary amplitude \(f(x)\) and the phase \(\phi(x)\) satisfy the following pair of equations:

\[-f''_{xx} + f(\phi'_x)^2 - f + f^3 = 0\]
\[f\phi''_{xx} + 2f'_x \phi'_x = -\beta f, \quad (3)\]

where the second equation is the continuity equation. At small \(\beta << 1\) one can neglect the second derivative of the stationary amplitude \(f''_{xx} \propto \beta^2\), so that \(\phi'_x = (1 - f^2)^{1/2} \equiv k\) and \(f'_x = -\beta f k/(3k^2 - 1)\), which substitution into Eq. (2) yields

\[-\eta''_{xx} - 2\eta'_x \left(ik - \frac{\beta k}{3k^2 - 1}\right) + (1 - k^2)(\eta + \eta^*) = (i - u) \eta \quad (4)\]

For a sufficiently long time, \(t \lesssim 1/\beta \gg 1\), a solution can be found in the form: \(\eta(x, t) = a \exp[i(\omega t - qx)] + b \exp[-i(\omega t - qx)]\), where coefficients \(a\) and \(b\) satisfy the generalized Bogoliubov equations,

\[(1 - k^2 - q^2 - 2qk - \frac{2i\beta k}{3k^2 - 1} + \omega + i\omega) a = (k^2 - 1) b,\]
\[(1 - k^2 - q^2 + 2qk - \frac{2i\beta k}{3k^2 - 1} - \omega + i\omega) b = (k^2 - 1) a,\]

with the eigenvalues,

\[\omega = 2kq \pm \sqrt{(1 - k^2 + q^2 - \frac{2i\beta k}{3k^2 - 1} + i\omega)^2 - (1 - k^2)^2}. \quad (5)\]

In the absence of any decay and dissipation we recover the familiar Bogoliubov spectrum of excitations, shifted by the current,

\[\omega^B_\pm = 2kq \pm \sqrt{q^4 + 2(1 - k^2)q^2}. \quad (6)\]

The decay \(\beta\) and the dissipation \(u\) lead to an imaginary part of \(\omega\), which is

\[\Im \omega = \pm \left(\mu \Im \omega^B_\pm - \frac{2\beta q k}{3k^2 - 1}\right) \sqrt{q^4 + 2(1 - k^2)q^2}. \quad (7)\]

for small \(u, \beta \ll 1\). The instability appears when \(\Im \omega\) is negative. In the absence of the decay (\(\beta = 0\)) , this condition corresponds to the negative \(\omega^B_\pm\), or the positive \(\omega^B_\pm\), which is the case if

\[4k^2 \geq q^2 + 2(1 - k^2). \quad (8)\]

Hence the uniform superflow with sufficiently small momentum, \(k \leq 1/\sqrt{3}\) is stable, while the higher momentum states with \(k > 1/\sqrt{3}\) are unstable. A finite decay \(\beta \neq 0\) shrinks the region of the uniform superflow to \(k < 1/\sqrt{3}\) as shown in Fig.1.
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