

Radiative dark–bright instability and the critical Casimir effect in DQW exciton condensates

T. Hakioglu^{a,b,*}, Ege Özgün^a

^a Department of Physics, Bilkent University, 06800 Ankara, Turkey

^b Institute of Theoretical and Applied Physics, 48740 Turunç, Muğla, Turkey

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ABSTRACT

It is already well known that radiative interband interaction in the excitonic normal liquid in semiconducting double quantum wells is responsible for a negligible splitting between the energies of the dark and bright excitons enabling us to consider a four fold spin degeneracy. This has also lead many workers to naively consider the same degeneracy in studying the condensate. On the other hand, the non-perturbative aspects of this interaction in the condensed phase, e.g. its consequences on the order parameter and the dark–bright mixture in the ground state have not been explored. In this work, we demonstrate that the ground state concentrations of the dark and the bright exciton condensates are dramatically different beyond a sharp interband coupling threshold where the contribution of the bright component in the ground state vanishes. This shows that the effect of the radiative interband interaction on the condensate is nonperturbative.

We also observe in the free energy a discontinuous derivative with respect to the layer separation at the entrance to the condensed phase, indicating a strong critical Casimir force. An estimate of its strength shows that it is measurable. Measuring the Casimir force is challenging, but at the same time it has a conclusive power about the presence of the long sought for condensed phase.

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The condensation of excitons in bulk semiconductors was speculated on a long time ago by Moskaleenko and Blatt [1,2]. A number of experiments have been performed since then, first in bulk systems, later in confined geometries using coupled quantum wells [3–6] with and without a strong magnetic field where the exciton lifetime can be increased by a factor of 10^3 – 10^4 (compared to that in bulk) allowing thermal equilibrium to be reached before recombination [7]. The exciton condensate primarily consists of fermionic pairs from *s*-like conduction and *p*-like valence bands. This pairing is fundamentally different from the conventional superconducting pairing [8] in that the four exciton states at sufficiently low temperatures are formed by *s*-like conduction electrons with an effective mass of $m_e^* \simeq 6.7 \times 10^{-2} m_e$ and *p*-like heavy holes $m_h^* \simeq 0.4 m_e$ with m_e the bare electron mass. The *p*-like bands are composed of light and heavy holes with the light holes being in a higher valence energy band than the heavy holes in 2-D geometries. This property has important consequences in the manifestation of the fermion exchange and time reversal symmetry operations [9] particularly in the presence

of a stabilizing electric field that is used in the experiments for confining the carriers in separate wells.

The experimental search for exciton condensate has been focused primarily on photoluminescence experiments [3–6] and after intense search for years in the confined geometries, unquestionable evidence for the condensed state is still lacking [7]. A proposition to resolve this dilemma has recently been made by Combescot and coworkers [10] stressing on the presence of radiative interband interactions. Due to the spin independent Coulomb interaction, it is naively expected that the excitons formed by the spin $\pm 1/2$ conduction electrons and the spin $\pm 3/2$ heavy holes form four degenerate spin configurations. According to the angular momentum selection rules however, the dark states composed of total spin ± 2 do not interact with the radiation field, whereas the bright states with spin ± 1 are coupled to it. This additional interaction of the bright states not only gives the bright pairs their shorter lifetime but also creates an effective interaction between the indirect electron–hole bands. In the quantum well geometries, these radiative processes are dipole like which can be made arbitrarily weak by a high tunneling barrier between the wells. These processes are already well known, in the excitonic normal liquid phase, to create a weak splitting between the dark and bright exciton lines, shifting the bright line slightly above in energy [11]. The same process, in the low temperature condensed phase, should therefore create an imbalance in the

* Corresponding author at: Department of Physics, Bilkent University, 06800 Ankara, Turkey. Tel.: +90 312 290 2109 (off.); fax: +90 312 266 4579.

E-mail address: hakioglu@bilkent.edu.tr (T. Hakioglu).

relative contributions of the dark and the bright components in favour of the dark excitons, which can make the ground state hardly visible in photoluminescence experiments.

In this work, we study the effect of the radiative interband processes on the condensed phase. In addition to the weak dark–bright splitting that is also present in the normal liquid, the breaking of the four-fold spin degeneracy to the two-fold Kramers' degeneracy results in a spin dependent instability in transition to the condensed phase. The original results of this work are that, when the interband radiation coupling exceeds a critical threshold, the bright condensate (BC) becomes sharply suppressed in the ground state whereas the dark condensate (DC) is enhanced implying that the effect is non-perturbative and the dark–bright balance in the ground state is strongly broken.

The radiative interband processes can be properly taken into account by including the dipole-field interaction that is present for the bright pairs. The radiation field can then be eliminated using the Markov–Lindblad formalism to obtain the reduced density matrix for the electrons and the holes, yielding an effective interaction of which the unitary part is an effective Hamiltonian for the bright states. We consider the condensate in the self consistent mean field Hartree–Fock scheme in the presence of this effective Hamiltonian.

In the absence of the radiative interband interaction, the Hamiltonian for the condensate is given in the electron–hole spinor basis $(\hat{e}_{\mathbf{k},\uparrow}, \hat{e}_{\mathbf{k},\downarrow}, \hat{h}_{-\mathbf{k},\uparrow}^\dagger, \hat{h}_{-\mathbf{k},\downarrow}^\dagger)$ by

$$\mathcal{H} = \begin{pmatrix} \tilde{\epsilon}_{\mathbf{k}}^{(x)} \sigma_0 & \Delta^\dagger(\mathbf{k}) \\ \Delta(\mathbf{k}) & -\tilde{\epsilon}_{\mathbf{k}}^{(x)} \sigma_0 \end{pmatrix} + \tilde{\epsilon}_{\mathbf{k}}^{(-)} \sigma_0 \otimes \sigma_0 \quad (1)$$

where $\sigma_0 \otimes \sigma_0$ is the 4×4 unit matrix, and in standard notation $\tilde{\epsilon}_{\mathbf{k}}^{(-)} = (\tilde{\epsilon}_{\mathbf{k}}^{(e)} - \tilde{\epsilon}_{\mathbf{k}}^{(h)})/2$, $\tilde{\epsilon}_{\mathbf{k}}^{(x)} = (\tilde{\epsilon}_{\mathbf{k}}^{(e)} + \tilde{\epsilon}_{\mathbf{k}}^{(h)})/2$ with

$$\tilde{\epsilon}_{\mathbf{k}}^{(p)} = \hbar^2 \mathbf{k}^2 (2m_p)^{-1} - \mu_p + \Sigma_{\mathbf{k}}^{(p)} \quad p = (e, h)$$

$$\Sigma_{\mathbf{k}}^{(p)} = \frac{1}{A} \sum_{\mathbf{q}} \mathcal{V}_{pp}(\mathbf{q}) \langle \hat{p}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{p}_{\mathbf{k}+\mathbf{q},\sigma} \rangle \quad (2)$$

$$\Delta_{\sigma\sigma'}(\mathbf{k}) = \frac{1}{A} \sum_{\mathbf{q}} \mathcal{V}_{eh}(\mathbf{q}) \langle \hat{e}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{h}_{-\mathbf{k}-\mathbf{q},\sigma'}^\dagger \rangle$$

the single particle energies, the spin dependent electron/hole self energies and the elements of the order parameter matrix $\Delta(\mathbf{k})$ respectively. The Coulomb interaction is given by $\mathcal{V}_{pp} = 2\pi e^2 (\epsilon q)^{-1}$ and $\mathcal{V}_{eh} = \mathcal{V}_{ee} e^{-qd}$ with d as the separation between the coupled quantum wells and the doubly degenerate spectrum of Eq. (1) is given by $E_{\mathbf{k}} = \tilde{\epsilon}_{\mathbf{k}}^{(-)} \pm [(\tilde{\epsilon}_{\mathbf{k}}^{(x)})^2 + \text{Tr}\{\Delta(\mathbf{k})\Delta^\dagger(\mathbf{k})\}/2]^{1/2}$. The time reversal symmetry is manifest and requires that $\Delta_{\sigma\sigma}(\mathbf{k}) = \Delta_{\sigma\sigma}^*(-\mathbf{k}) = \Delta_D(\mathbf{k})$ and $\Delta_{\sigma\bar{\sigma}}(\mathbf{k}) = -\Delta_{\sigma\bar{\sigma}}^*(-\mathbf{k}) = \Delta_B(\mathbf{k})$ corresponding to the dark (D) and the bright (B) components of the condensate. The spin independence of the Coulomb interaction requires that $|\Delta_D(\mathbf{k})| = |\Delta_B(\mathbf{k})|$. The analytical solution of this problem can be formulated within the Hartree–Fock mean field theory yielding two doubly degenerate exciton branches due to the underlying spin degeneracy and Kramers' symmetry. On the other hand, the spin degeneracy is broken by the interband radiation field with only the two fold Kramers' symmetry remaining. The bright excitons couple to the radiation field by the dipole coupling

$$\mathcal{H}_{\text{rad}} = - \sum_{\mathbf{q}} \mathbf{p}(\mathbf{q}) \cdot \mathbf{E}(\mathbf{q}) \quad (3)$$

where $\mathbf{p}(\mathbf{q})$ is the bright exciton dipole moment

$$\mathbf{p}(\mathbf{q}) = \sum_{\mathbf{k},\sigma} \mathbf{p}_0 \hat{e}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{h}_{-\mathbf{k},\sigma}^\dagger + \text{h.c.} \quad (4)$$

with $\mathbf{p}_0 = \int d\mathbf{r} \Psi_e(\mathbf{r}) \mathbf{r} \Psi_h(\mathbf{r})$ the dipole matrix element depending on the overlap of the electron and hole orbitals and $\mathbf{E}(\mathbf{q})$ is the quantized electric field. We consider

$$\mathbf{p}_0 = e d e^{-d^2/W^2} \mathbf{e}_z \quad (5)$$

where e is the elementary charge, \mathbf{e}_z is the unit vector perpendicular to the quantum well plane, and $d \simeq 100 \text{ \AA}$ and $W \simeq 70 \text{ \AA}$ are the typical layer separation and the well width respectively. Eq. (4) is spin anti-correlated between the electron–hole spins; hence \mathcal{H}_{rad} in Eq. (3) is only present for the bright states. In terms of the quantized radiation field, Eq. (3) is

$$\mathcal{H}_{\text{rad}} = \frac{-i}{\sqrt{V}} \sum_{\substack{\mathbf{q},\lambda \\ (\sigma,\lambda)}} \alpha_{\mathbf{q},\lambda} (\hat{S}_{\mathbf{k},\mathbf{q},\sigma}^\dagger + \hat{S}_{\mathbf{k},-\mathbf{q},\sigma}) (\hat{a}_{\mathbf{q},\lambda} - \hat{a}_{-\mathbf{q},\lambda}^\dagger) \quad (6)$$

where $\hat{S}_{\mathbf{k},\mathbf{q},\sigma} = \hat{e}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{h}_{-\mathbf{k},\sigma}^\dagger$ is the exciton creation operator, $a_{\mathbf{q},\lambda}/a_{\mathbf{q},\lambda}^\dagger$ are the radiation field annihilation/creation operators and $\alpha_{\mathbf{q},\lambda} = \sqrt{2\pi \hbar \omega_{\mathbf{q}}/\epsilon} \mathbf{p}_0 \cdot \boldsymbol{\xi}_{\mathbf{q},\lambda}$ is the coupling strength with $\hbar \omega_{\mathbf{q}}$, and $\boldsymbol{\xi}_{\mathbf{q},\lambda}$ being the photon energy and the polarization vector with λ as the polarization branch index. The static dielectric constant of the medium is $\epsilon \simeq 12\epsilon_0$ with ϵ_0 being that of the vacuum. We derive the equation of motion for the full density matrix including the second order terms in $\alpha_{\mathbf{q},\lambda}$. Using the Markov–Linblad formalism to trace out the radiation field, we find an effective Hamiltonian for the reduced density matrix for the electrons and holes. This effective Hamiltonian is second order in $\alpha_{\mathbf{q},\lambda}$ with unitary and non unitary parts; with the former yielding the effective interaction, and the latter the finite lifetime corrections. We study the unitary contribution given by

$$\mathcal{H}_e = - \sum_{\substack{\mathbf{q},\lambda \\ (\sigma,\sigma')}} \left(\Omega_{\mathbf{k},\mathbf{q}}^{(+)} \hat{S}_{\mathbf{k},\mathbf{q},\sigma}^\dagger \hat{S}_{\mathbf{k},\mathbf{q},\sigma'} + \Omega_{\mathbf{k},\mathbf{q}}^{(-)} \hat{S}_{\mathbf{k},\mathbf{q},\sigma} \hat{S}_{\mathbf{k},\mathbf{q},\sigma'}^\dagger \right) \quad (7)$$

where

$$\Omega_{\mathbf{k},\mathbf{q}}^{(\pm)} = |\alpha_{\mathbf{q}}|^2 \text{Im}\{\tilde{F}_{\mathbf{q}}(\epsilon_{\mathbf{k},\mathbf{q}})\} \begin{pmatrix} \left(1 - \frac{\sin^2 \theta_{\mathbf{k}}}{2}\right) \cos^4(\theta_{\mathbf{k}}/2) \\ \left(1 - \frac{\sin^2 \theta_{\mathbf{k}}}{2}\right) \sin^4(\theta_{\mathbf{k}}/2) \end{pmatrix} \quad (8)$$

is the effective coupling strength of the radiation field in the presence of the condensate with $\epsilon_{\mathbf{k},\mathbf{q}} = \tilde{\epsilon}_{\mathbf{k}+\mathbf{q}}^{(e)} - \tilde{\epsilon}_{\mathbf{k}}^{(h)}$, $|\alpha_{\mathbf{q}}|^2 = \sum_{\lambda} |\alpha_{\mathbf{q},\lambda}|^2 = 2\pi \hbar \omega_{\mathbf{q}} \epsilon^{-1} (p_0^2 - |\mathbf{p}_0 \cdot \mathbf{q}|^2/q^2)$ and $\tilde{F}_{\mathbf{q}}(\omega) = \hbar^{-1} \int_0^\infty dt e^{i\omega t} \langle A_{\mathbf{q}}(t) A_{-\mathbf{q}}(0) \rangle$ where $A_{\mathbf{q}}(t) = (\hat{a}_{\mathbf{q}} - \hat{a}_{-\mathbf{q}}^\dagger)$ with the spectral function of the radiation field where $\tilde{F}_{\mathbf{q}}(\omega) = i\mathcal{P}[1/(\omega - \hbar \omega_{\mathbf{q}})] + \delta(\omega - \hbar \omega_{\mathbf{q}})$ with \mathcal{P} standing for the principle value. In Eq. (8) $\sin^2 \theta_{\mathbf{k}} = [\Delta_D^2(\mathbf{k}) + \Delta_B^2(\mathbf{k})]/E_{\mathbf{k}}^2$ is the coherence factor of the condensate. We simplify the effective interaction in Eq. (7) by ignoring the contribution of the small radiation field momentum in comparison with the electron and hole momenta. In the mean field approximation the effective Hamiltonian is

$$\mathcal{H}_e \simeq - \sum_{\mathbf{k},\sigma,\sigma'} \left[g_{\mathbf{k}} \langle \hat{e}_{\mathbf{k},\sigma}^\dagger \hat{h}_{-\mathbf{k},\sigma}^\dagger \rangle \hat{h}_{-\mathbf{k},\sigma'} \hat{e}_{\mathbf{k},\sigma'} + \text{h.c.} \right] \quad (9)$$

where, using Eq. (8) and the definitions thereafter,

$$g_{\mathbf{k}} = \sum_{\mathbf{q}} (\Omega_{\mathbf{k},\mathbf{q}}^{(+)} + \Omega_{\mathbf{k},\mathbf{q}}^{(-)}) = \left(1 - \frac{\sin^2 \theta_{\mathbf{k}}}{2}\right)^2 \sum_{\mathbf{q}} |\alpha_{\mathbf{q}}|^2 \left[\frac{\epsilon_{\mathbf{k},\mathbf{q}} - \hbar \omega_{\mathbf{q}}}{(\epsilon_{\mathbf{k},\mathbf{q}} - \hbar \omega_{\mathbf{q}})^2 + \eta^2} \right] \quad (10)$$

where η is infinitesimal. In calculating Eq. (10) we neglect the quadratic terms in \mathbf{q} and use the fact that \mathbf{q} is largely confined to the double well plane, e.g. $|\mathbf{p}_0, \mathbf{q}| \ll p_0 q$, yielding

$$\frac{g_{\mathbf{k}}}{g_0} = - \left[1 - \frac{1}{2} \frac{\Delta_D^2(\mathbf{k}) + \Delta_B^2(\mathbf{k})}{E_{\mathbf{k}}^2} \right]^2 f(x), \quad g_0 = \frac{2}{\pi} \frac{q_{\max}^2 p_0^2}{\epsilon d} \quad (11)$$

where $x = \hbar^2 k^2 / (m_x^2 c^2)$ and $f(x) \simeq 1$ for $x \ll 1$. Here, q_{\max} is the maximum photon wavevector which is estimated as $q_{\max} = E_G / (\hbar c) = 5 \times 10^{-4} \text{ \AA}^{-1}$ with $E_G \simeq 1 \text{ eV}$ as the semiconductor band gap. With these values the overall physical energy scale in Eq. (11) becomes $g_0^{(p)} \simeq 10^{-2} \text{ meV}$. The most important effect of this small contribution is not the dark–bright energy shift, but breaking the four fold spin degeneracy of the effective electron–hole interaction. Inserting Eq. (9) into Eq. (1) we obtain a model Hamiltonian which can be solved exactly. The order parameter is now different for the dark and the bright states as given by

$$\tilde{\Delta}_{\sigma\sigma'}(\mathbf{k}) = \frac{1}{A} \sum_{\mathbf{q}} [\mathcal{V}_{eh}(\mathbf{q}) - \delta_{\sigma',\bar{\sigma}} \delta_{\mathbf{q},0} g_{\mathbf{k}}] \langle \hat{c}_{\mathbf{k}+\mathbf{q},\sigma}^\dagger \hat{h}_{-\mathbf{k}-\mathbf{q},\sigma'}^\dagger \rangle \quad (12)$$

with $\tilde{\Delta}_{\sigma\bar{\sigma}}(\mathbf{k}) \neq \tilde{\Delta}_{\sigma\sigma}(\mathbf{k}) = \Delta_{\sigma\sigma}(\mathbf{k})$, i.e. the dark components are unaffected. The analytic solution for the order parameter is a coupled set of equations for the dark and bright components given by,

$$\Delta_D(\mathbf{k}) = \frac{1}{2A} \sum_{\mathbf{k}'} \mathcal{V}_{eh}(\mathbf{k} - \mathbf{k}') \frac{\Delta_D(\mathbf{k}') [E_{\mathbf{k}'} + \tilde{\epsilon}_{\mathbf{k}'}^{(x)}]}{D_{\mathbf{k}'}} \times \left\{ [f_1(\mathbf{k}') + f_2(\mathbf{k}')] - [f_3(-\mathbf{k}') + f_4(-\mathbf{k}')] \right\} \quad (13)$$

$$\Delta_B(\mathbf{k}) = \frac{1}{2A} \sum_{\mathbf{k}'} \mathcal{V}_{eh}(\mathbf{k} - \mathbf{k}') \frac{\tilde{\Delta}_B(\mathbf{k}') [E_{\mathbf{k}'} + \tilde{\epsilon}_{\mathbf{k}'}^{(x)}]}{D_{\mathbf{k}'}} \times \left\{ [f_1(\mathbf{k}') + f_2(\mathbf{k}')] - [f_3(-\mathbf{k}') + f_4(-\mathbf{k}')] \right\}. \quad (14)$$

With two other equations for the electron and hole chemical potentials, these closed sets of equations are solved numerically. Eqs. (13) and (14) are not identical when $g_{\mathbf{k}} \neq 0$. Their numerical solutions are depicted in Fig. 1 as g_0 and the temperatures are varied. For $g_0 < 10^{-6} \text{ meV}$, the solutions are basically spin independent and the dark and the bright solutions are identical. Within a short interval in g_0 beyond this coupling, the DC becomes slightly stronger without a change in its temperature behaviour, whereas the BC is weakened by non-zero temperatures and it is completely absent for $g_0 > 10^{-6} \text{ meV}$. Considering that in most experiments $g_0 \simeq 10^{-2} \text{ meV} \simeq 10^{-3} E_H$, we observe that the bright state should not be present. The theory thus predicts that the DC should dominate the ground state. The phase boundary $d_c(n_x, n_-)$ beyond which the condensate completely disappears is shown in Fig. 2 for the DC. The hole like imbalance, i.e. $n_- < 0$, is preferable for the condensate compared to the electron like one, i.e. $0 < n_-$, due to the difference in the effective masses and this asymmetry, because it is a kinetic energy effect, is more pronounced when the exciton density is higher. We observed that in the range $1.5a_B < d_c < 3a_B$ as the exciton concentration is increased from $n_x = 0.5a_B^{-2}$ to $2a_B^{-2}$, d_c is nearly independent of n_x for a fixed n_- , whereas, for a fixed n_x the number imbalance in favour of holes increases d_c .

We report another important result which is the discontinuity observed in the derivative of the energy gap with respect to the layer separation, i.e. $\partial|\Delta(\mathbf{k})|/\partial d$. It can be found very simply that this quantity is directly related to the critical Casimir force [12]. It is given by $f_{\text{Cas}} = -\partial(\delta F)/\partial d$ which becomes for $d \simeq d_c$, $f_{\text{Cas}} \simeq \sum_{\mathbf{k}} (\partial \Delta^2(\mathbf{k})/\partial d) / (2|\tilde{\epsilon}_{\mathbf{k}}^{(x)}|) < 0$ where δF is the Helmholtz free energy of the condensate. We plot the maximum value of $|\Delta(\mathbf{k})|$ (at

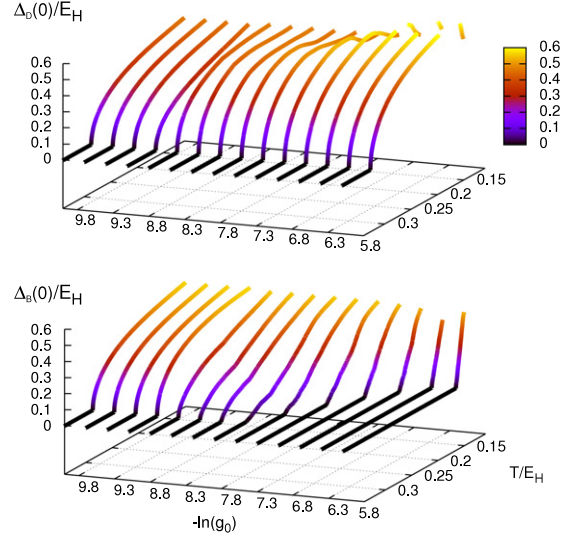


Fig. 1. (Colour online) DC and BC order parameters versus the radiative coupling strength (in log scale) and the temperature. The symmetric solution breaks abruptly at $g_0 \simeq 10^{-8} \text{ eV} \simeq 1.6 \times 10^{-6} E_H$ above which the DC (upper) is nearly stable with a robust critical temperature whereas the BC (lower) is rapidly suppressed. Here $E_H \simeq 6 \text{ meV}$ is a typical Hartree energy used as an energy scale.

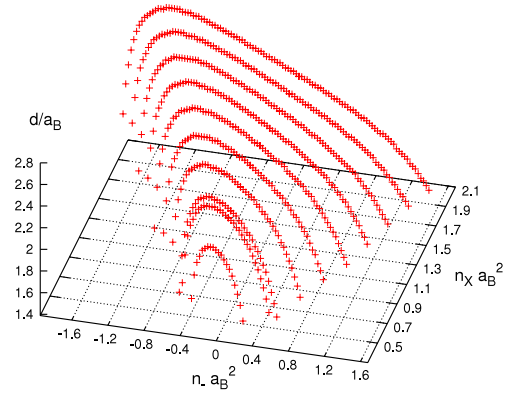


Fig. 2. The phase boundary $d_c(n_x, n_-)$. The DC order parameter vanishes for $d < d_c$ for a fixed n_x and n_- and the condensate is non zero inside the surface. The bright state is not condensed due to the high radiative coupling, i.e. $g_0 \simeq 10^{-3} E_H$. The asymmetry on the bounding surface is more pronounced for larger exciton numbers.

$\mathbf{k} = 0$) in Fig. 3 with respect to d and n_- which has a discontinuity at $d = d_c(n_x, n_-)$. $|\Delta(\mathbf{k} = 0)|$ is plotted in the inset of Fig. 3] as a function of n_- for $n_x a_B^2 = 1.2$. We observe that $|f_{\text{Cas}}|$ is the largest at the onset of condensation. We numerically estimate $|f_{\text{Cas}}| = (1 - 5) \times 10^{-1} E_H / a_B^* \simeq 10^{-13} \text{ N}$ which is, in principal, measurable [13]. The measurement of the critical Casimir force, although challenging, may thus be a strong signal for the presence of the exciton condensate with no known alternative explanation.

In conclusion, we have shown that the relative ground state concentrations of the DC and BC spinor components, are dramatically different due the radiative interband transitions, breaking the four fold spin degeneracy to the two fold Kramers' degeneracy. To our best knowledge, this work on the implications of the broken dark–bright symmetry is the only concrete result so far in the condensed ground state. In reality, some small concentration of the BC should be present in the ground state. The basic reason for this is the Shiva diagrams [10,14] produced by the exciton–exciton interactions where two dark excitons can turn into two bright ones and vice versa. In equilibrium, the relative concentrations of the dark and bright components are then

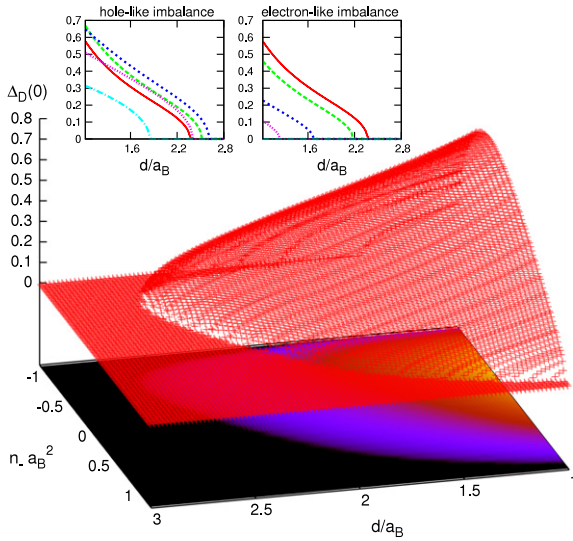


Fig. 3. (Colour online) DC order parameter as a function of layer separation d (in units of a_B^2) and n_- (in units of a_B^2) for $n_+ a_B^2 = 0.8$. The insets above are the cross sections of the surface below for hole-like imbalance (left), i.e. $n_- < 0$, and the electron-like imbalance (right), i.e. $0 < n_-$. The colours depict: red ($n_- = 0$), green ($|n_-| = 0.3$), blue ($|n_-| = 0.8$), purple ($|n_-| = 1.0$), and turquoise ($|n_-| = 1.14$).

dictated by the detailed balance which is expected to yield a non zero BC in the ground state.

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