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Quatron-polaritons: charged quasi-particles having the bosonic statistics

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Abstract

Negatively charged quatrons are quasi-particles composed of a hole and three electrons in a semiconductor. In bulk semiconductors they are unstable. We show that they become stable if coupled to light in specially designed triple quantum layer structures embedded in microcavities. Due to an extremely small effective mass, charged quatron-polaritons can form a superfluid at high temperatures. This opens up a breathtaking perspective on optically induced and controlled superconductivity at room temperature.

(Some figures in this article are in colour only in the electronic version)

Three sorts of stable electronic quasi-particles in semiconductors are well known: the electron, the hole, and the exciton. In addition, at low temperatures, the trion [1] (the composite of two electrons and one hole) and composite magnetic-flux bosons [2] (such as seen in the quantum Hall effect) have been observed at liquid helium temperatures and below. Anion excitons have been predicted in the quantum Hall regime as well [3]. Here we propose a new quasi-particle: the quatron. Negatively charged quatrons are composed of a hole and three electrons in a semiconductor. In bulk semiconductors they are unstable because of Coulomb repulsion of the electrons. We show that they become stable if coupled to light in specially designed triple quantum layer structures embedded in microcavities. Because quatrons are optically accessible bosons, they offer an entirely new possibility for Bose–Einstein condensation (BEC) in semiconductors, which we show can be robust up to room temperature.

The achievement of superconductivity at room temperature has been a challenging objective for physicists for many years. Despite the optimism inspired by the discovery of high-temperature superconductors in the 1980s, this objective is still far from being achieved [4]. According to the Bardeen–Cooper–Schrieffer (BCS) model, superconductivity requires the pairing of charge carriers. A Cooper pair of electrons has an integer spin and therefore exhibits bosonic properties. Superfluidity of electronic pairs leads to superconductivity [5].

In the BCS model the pairing is caused by electron–phonon interaction. The critical temperature in BCS superconductors remains lower than 200 K in all materials studied up to now. The exciton mechanism for superconductivity, which was proposed a long time ago

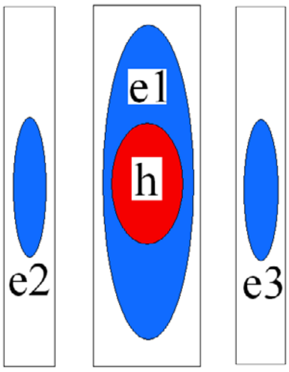

Figure 1. A sketch of the quatron state in a triple quantum layer.

(see e.g. [6]), is based on coupling of the electrons from a metal with a virtual exciton in a semiconductor and the eventual formation of a Cooper pair. This mechanism does not rely on the BEC of excitons and remains essentially within the BCS model, while assuming a different pairing mechanism. It can hardly give a strong increase in the critical temperature of the superfluid phase transition and has never been observed experimentally.

We propose an alternative mechanism for pairing of electrons which can allow, in principle, superfluidity of charged bosons at high temperatures. In our model, a pair of conductivity electrons is linked with an exciton-polariton [7] by Coulomb interaction. As a result, a complex quasi-particle with a charge of $Q = -2e$ appears. This can be called a quatron, by analogy with the well-known trion (a complex of one hole and two electrons). The quatrons are unstable in bulk semiconductors because of Coulomb repulsion between electrons. Moreover, the quatrons have been shown to form no discrete quantum-confined states in quantum wells (QWs) containing a two-dimensional electron gas (2DEG) [8, 9], where they only contribute to broadening of the trion and exciton states. Reference [8] explicitly states that quatrons '. . . do not lead to a new bound state with an energy lower than the trion but broaden both the exciton and the trion line'.

Here we show that the quatrons may become stable and form discrete localized states due to coupling with optical modes in specially designed microcavity structures. We propose to embed in the conventional planar microcavity a structure favourable for the formation of quatrons that consists of three QWs. The largest QW is in the centre. It is undoped and possesses an excitonic transition at the energy E_x. Two side QWs are thinner, so that they are optically transparent at the energy E_x. They contain a shallow 2DEG created by n-doping of the outer barriers. A quatron is formed if two electrons from the side wells are attached to the exciton in the central layer. The advantage of a three-QW structure with respect to a single QW is evident: exchange coupling between spatially separated electrons is strongly reduced; moreover, the central well does not contain the 2DEG, thus the excitonic transition in this well is not weakened due to the phase-space filling effect. We find that, even in such a favourable system, the stability of quatrons requires a strong difference between the effective masses of electrons in the central and side wells and strong exciton–photon coupling. The electron in the central well should be light enough to provide an exciton Bohr radius that is larger or comparable to the interwell spacing, while side-band electrons should be heavy enough to correlate strongly their in-plane motion with the hole (see figure 1).

The above-mentioned conditions are met in the GaAs/AlAs/$Al_{0.45}Ga_{0.65}As$ structure shown schematically in figure 2. A thin GaAs QW is also confined by thin AlAs barriers.

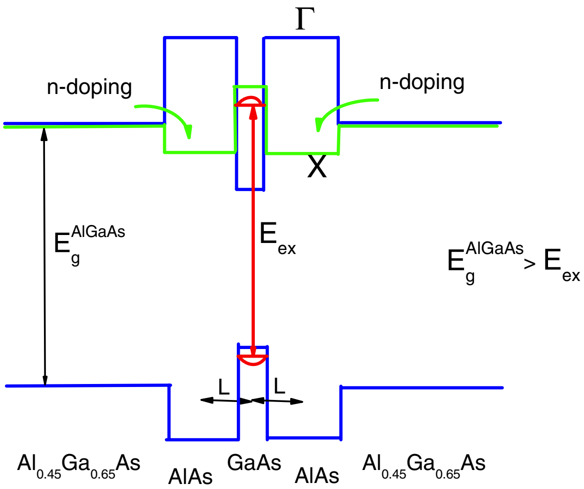

Figure 2. Band diagram of the structure proposed for detection of quatron-polaritons in microcavities.

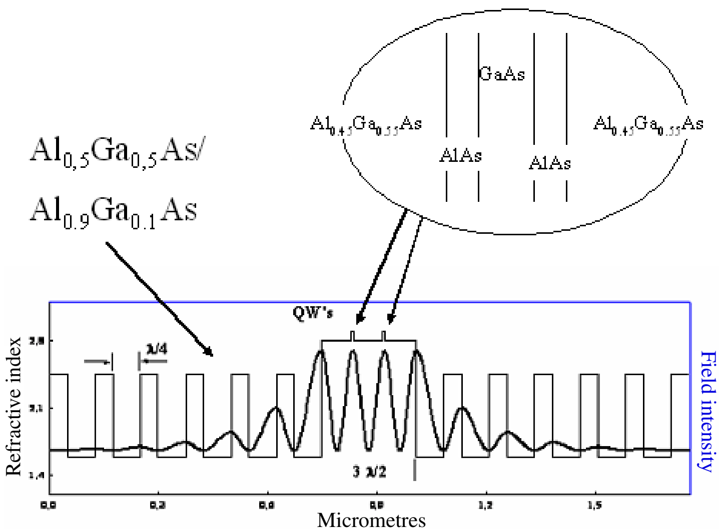

Figure 3. Microcavity structure suitable for observation of quatron polaritons.

These barriers represent potential wells for X-valley electrons, which have much larger effective masses than Γ-valley electrons from the central well.

These side wells are populated by a shallow 2DEG due to n-doping of the surrounding AlGaAs barriers or due to optical excitation of the electron–hole pairs in the barriers (in this case, the electrons get trapped into X-valleys of the AlAs layers due to the ΓX mixing, while the holes remain outside). Electronic Γ and X-levels in the surrounding layers of AlGaAs are lower in energy than the lowest Γ-electron level in the central QW, so that penetration of the electrons into the central well by tunnelling is excluded. Layer thicknesses are chosen so that the lowest-energy allowed optical transition in the system is associated with the Γ-exciton confined in the central well.

In order to take advantage of strong coupling with light, the above structure should be embedded in a standard microcavity, as shown schematically in figure 3. We have developed a variational procedure that allows one to estimate the energy of the quatron-polariton ground state in a microcavity.

Let L be the distance between centres of quantum-confined layers, $\vec{\rho}_1 = \vec{\rho}_{\rm h} - \vec{\rho}_{\rm e1}$, $\vec{\rho}_2 = \vec{\rho}_{\rm h} - \vec{\rho}_{\rm e2}$ and $\vec{\rho}_3 = \vec{\rho}_{\rm h} - \vec{\rho}_{\rm e3}$, where $\vec{\rho}_{\rm h}$ and $\vec{\rho}_{\rm e1}$ are radius vectors of the hole and the electron in the central layer, $\vec{\rho}_2$ and $\vec{\rho}_3$ are radius vectors of the electrons in the side-wells, and $\mu_1 = \frac{m_{\rm e1}m_{\rm h}}{m_{\rm e1}+m_{\rm h}}$, $\mu_2 = \frac{m_{\rm e2}m_{\rm h}}{m_{\rm e2}} + m_{\rm h}$, $m_{\rm h}$, $m_{\rm e1}$ and $m_{\rm e2}$ are effective masses of the hole and electron in the central layer and the electron in the side layer. We consider the following Hamiltonian describing the quatron-polariton state:

$$\hat{H} = \begin{bmatrix} \hat{H}_{\rm coul} & V\delta(\rho_1) \\ V\delta(\rho_1) & \hbar\omega_{\rm c} \end{bmatrix}, \tag{1}$$

where $\hbar\omega_{\rm c}$ is the energy of the bare cavity mode,

$$\begin{aligned} \hat{H}_{\rm coul} = &- \sum_{i=1,2,3} \frac{\hbar^2}{2\mu_i}\frac{1}{\rho_i}\frac{\partial}{\partial\rho_i}\left(\rho_i\frac{\partial}{\partial\rho_i}\right) - \frac{e^2}{\varepsilon\rho_1} - \frac{e^2}{\varepsilon\sqrt{L^2+\rho_2^2}} - \frac{e^2}{\varepsilon\sqrt{L^2+\rho_3^2}} \\ &+ \frac{e^2}{\varepsilon\sqrt{L^2+\rho_1^2+\rho_2^2-2\rho_1\rho_2\cos(\vec{\rho}_1,\vec{\rho}_2)}} \\ &+ \frac{e^2}{\varepsilon\sqrt{L^2+\rho_1^2+\rho_3^2-2\rho_1\rho_3\cos(\vec{\rho}_1,\vec{\rho}_3)}} \\ &+ \frac{e^2}{\varepsilon\sqrt{(2L)^2+\rho_3^2+\rho_2^2-2\rho_3\rho_2\cos(\vec{\rho}_3,\vec{\rho}_2)}}. \end{aligned} \tag{2}$$

The matrix element $\mathbf{V}$ describes the exciton–light coupling. The extension of the exciton and electron wavefunctions in the direction normal to the QW plane is neglected here, which is a reasonable assumption for thin quantum wells.

The trial function is a two-dimensional vector of the form

$$\begin{bmatrix} \alpha\Psi(\rho_1,\rho_2,\rho_3) \\ \beta \end{bmatrix}, \tag{3}$$

where

$$\Psi(\rho_1,\rho_2,\rho_3) = \left(\frac{2}{\pi}\right)^{3/2} \frac{\exp\left[\frac{2L}{a_2}\right]}{a_1 a_2(a_2+2L)} \exp\left[-\frac{\rho_1}{a_1} - \frac{\sqrt{\rho_2^2+L^2}+\sqrt{\rho_3^2+L^2}}{a_2}\right]. \tag{4}$$

At the anti-crossing point, one can fix $\alpha = -\frac{\beta}{\sqrt{S}} = \frac{\sqrt{2}}{2}$ (where S is the surface area) and use as variational parameters only a_1 and a_2. In this case the exciton-polariton energy is given by

$$E_{\rm ex} - \hbar\omega_{\rm c} = \frac{\hbar^2}{2\mu_1 a_1^2} - \frac{2e^2}{\varepsilon a_1} - V\frac{\hbar^2\varepsilon}{2\mu_1 e^2 a_1}, \tag{5}$$

where $2V$ is the Rabi splitting. This should be minimized with respect to a_1.

The energy of the quatron-polariton is given by:

$$E - \hbar\omega_{\rm c} = \frac{\hbar^2}{2\mu_1 a_1^2} - \frac{2e^2}{\varepsilon a_1} - V\frac{\hbar^2\varepsilon}{2\mu_1 e^2 a_1} + T_2 - \frac{4e^2}{\varepsilon(a_2+2L)}$$

$$+\frac{16e^2\exp\left[\frac{2L}{a_2}\right]}{\pi\varepsilon a_1^2 a_2\left(a_2+2L\right)}\int_0^{2\pi}\mathrm{d}\varphi\int_0^{\infty}\int_0^{\infty}\rho_1\,\rho_2$$

$$\times\frac{\exp\left[-2\frac{\sqrt{\rho_2^2+L^2}}{a_2}-2\frac{\rho_1}{a_1}\right]}{\sqrt{L^2+\rho_1^2+\rho_2^2-2\rho_1\rho_2\cos\varphi}}\mathrm{d}\rho_1\mathrm{d}\rho_2$$

$$+\frac{8e^2}{\pi\varepsilon}\frac{\exp\left[\frac{4L}{a_2}\right]}{a_2^2\left(a_2+2L\right)^2}\int_0^{2\pi}\mathrm{d}\varphi\int_0^{\infty}\int_0^{\infty}\rho_3\,\rho_2$$

$$\times\frac{\exp\left[-2\frac{\sqrt{\rho_2^2+L^2}}{a_2}-2\frac{\sqrt{\rho_3^2+L^2}}{a_2}\right]}{\sqrt{4L^2+\rho_3^2+\rho_2^2-2\rho_3\rho_2\cos\varphi}}\mathrm{d}\rho_3\,\mathrm{d}\rho_2, \quad (6)$$

where

$$T_2=\frac{2\hbar^2\exp\left[\frac{2L}{a_2}\right]}{\mu_2 a_2^3\left(a_2+2L\right)}\int_{L^2}^{\infty}\mathrm{d}x\mathrm{e}^{-\frac{2\sqrt{x}}{a_2}}\frac{a_2\left(x+L_2^2\right)-\sqrt{x}\left(x-L^2\right)}{x^{3/2}}. \quad (7)$$

Here we have neglected the exchange terms, as they give only a minor contribution if the confinement of electrons in separate quantum layers is strong enough.

The quatron energy should be minimized with respect to a_1 and a_2 and, if the resulting value is less than E_{ex}, the quatron-polariton is stable. Figure 4(a) shows the calculated dependences of exciton-polariton and quatron-polariton energies on the distance L between the centres of the GaAs and AlAs layers. We have used the following set of parameters: $\varepsilon = 13$, $m_{\mathrm{e1}} = 0.063\,m_0$, $m_{\mathrm{e2}} = 0.22\,m_0$ and $V = 3.5$ meV. One can see that the quatron-polariton is stable in a wide range of parameters and that its binding energy increases with a decrease in L. This increase is accompanied by a decrease in the effective Bohr radius, a_2, which determines the in-plane size of the quatron. In real systems this tendency is limited due to penetration of electron wavefunctions into the barriers. Interestingly, coupling with the microcavity mode makes the quatrons more stable. Effectively, the quatron-polariton energy is shifted down with respect to the bare quatron energy by a half of the Rabi-splitting. The Rabi-splitting changes proportionally to the inverse Bohr radius $1/a_1$, thus stronger localized states have lower energy, while dissociation leads to significant energy losses. The effect of strong optical coupling on the exciton Bohr radius has been pointed out by Khurgin [10]. Figure 4(c) shows the dependence of the binding energy of the quatron-polariton of the exciton–light coupling constant V. The stability of quatron-polaritons decreases with a decrease in the strength of their interaction with light. When V reaches a very small value, the quatron becomes unstable. These results show that, at least for the set of parameters that we used, a 'bare' quatron is unstable whereas a quatron-polariton is a stable quasi-particle.

We have also checked that a single QW containing a 2DEG would not have a stable quatron state even if it is embedded in the microcavity. The reason lies in the strong exchange interaction between electrons, which leads to an increase in the quatron energy in this case. This calculation has been performed with the Hamiltonian (2) at $L = 0$. In order to minimize the negative effect of electron–electron exchange interaction, we have taken either 1s, 2s, 2p or 2s, $2\mathrm{p}_x$, $2\mathrm{p}_y$ trial functions for the relative motion of the hole and three electrons. Both combinations yield a quatron-polariton energy that exceeds the trion energy. We realize that the variational approach is not a good one if the goal is to show that the given state is unstable, as it always yields a higher energy than the exact solution. The check that we performed is still important, as it shows that there is no contradiction between our work and [8]: under the

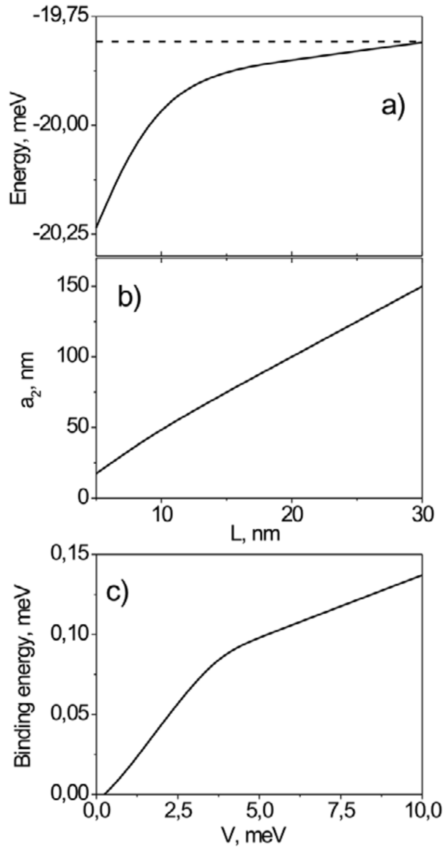

Figure 4. Results of the variational calculation of the energy of a quatron-polariton (solid) as a function of the spacing L between the centres of potential wells for Γ and X-electrons. The dashed line shows the exciton-polariton energy (a). Zero at the vertical axis would correspond to the uncoupled free exciton energy. Variation of the effective in-plane radius of the quatron as a function of L (b), and dependence of the quatron-polariton binding energy on the exciton–photon coupling strength V calculated for $L = 10$ nm (c).

same conditions, both works lead to the same conclusion. The original result of this work is that in different conditions (a triple quantum layer in a microcavity) the quatron-polaritons can be stable.

Though the quatron-polariton is stable in a microcavity, its energy difference from an exciton-polariton is small and the quatron-polariton is expected to be ionized at temperatures of the order of 4 K in realistic systems. However, if Bose condensed, the quatron-polaritons become much more robust, as in this case their binding energy multiplied by the population of the condensate should be compared with the temperature. Stability of quatron-polariton superfluids is given by the condition

$$N\delta E \gg k_{\mathrm{B}}T, \tag{8}$$

where N is the population of the condensate, δE is the difference between the quatron-polariton and exciton-polariton energies, and T is the temperature. The condition (8) can be easily understood in terms of the BEC in a two-level system. The energy levels corresponding to the charged quatron-polariton and neutral exciton polariton states are split by δE. In the limit of large N, the condensate can be considered as a single classical spin having $N + 1$ allowed projections to an effective magnetic field axis. The energies of the system corresponding to the different spin projections form an equidistant ladder and range from $-\frac{N}{2}\delta E$ to $\frac{N}{2}\delta E$. In order to flip the spin (i.e. to move the condensate from a purely quatron-polariton to a purely exciton-polariton state) one needs to spend an energy equal to $N\delta E$.

We underline here that no significant spectral features associated with trions (two electron and one hole) can be expected if the polariton condensate is formed, for the simple reason that trions, being fermions, obey the Pauli principle, and only a single polariton is allowed to occupy a given trion quantum state[3]. On the other hand, quatron-polaritons, being bosons, can be subject to condensation and superfluidity. The spin of a stable quatron is equal to zero (singlet state), which is achieved by combining an optically active exciton having a spin ± 1 with a pair of electrons having a total spin of ∓ 1.[4] The spatial separation of excitons from the electrons will also help to avoid exciton bleaching by the electron gas and oscillator strength losses due to phase-space filling.

Bose–Einstein condensation of excitons has been discussed in the literature since the 1960s, but still no convincing experimental observation of it has been reported [11]. The reason is that excitons are subject to various dephasing mechanisms, they are sensitive to all kinds of localizing potentials, and have a too heavy effective mass to be condensed at temperatures higher than the liquid helium temperature. On the other hand, exciton-polaritons formed by excitons coupled with photon modes in microcavities can be condensed much more easily and at much higher temperatures, according to theoretical predictions and preliminary experimental results [11–15]. The exciton-polaritons in microcavities have an in-plane effective mass that is four orders of magnitude less than a typical exciton effective mass and eight orders of magnitude lighter than the He atom mass [7]. This makes them extremely good candidates for Bose condensation at room temperature even though, due to extremely short radiative lifetime, the polaritons can hardly achieve thermal equilibrium in microcavities [16]. Strong coupling with light also prevents the locaization of excitons on structural imperfections: the polariton state has a coherence length of the order of the wavelength of light, i.e. much larger than the typical disorder scale.

Exciton-polaritons are neutral, therefore they cannot give any contribution to conductivity. However, bound to electron pairs they may form a charged superfluid. This superfluid will survive whilst the microcavity is optically pumped. This is why we deal with optically induced superfluidity of charged particles. Scattering with free electrons could lead to ionization of the quatron-polaritons, so one should be able to control effectively the concentration of free electrons in order to have them close to the concentration of polaritons in the condensate. This is why optical creation of electron–hole pairs seems to us to be strongly advantageous: by changing the pumping power, one can tune the density of the electrons trapped into the AlAs layers to the condensate density. We must underline that the GaAs/AlAs/AlGaAs system considered here would not allow observation of superfluidity at room temperature, as the exciton binding energy in GaAs is too small. In order to study quatron-polaritons at higher temperatures, one should use similar structures but based on wide-band-gap semiconductor materials such as GaN or ZnO.

To summarize, the *concept of quatron-polariton superfluidity/superconductivity*is based on the following statements:

- in carefully designed structures, quatron-polaritons have lower energies than exciton-polaritons, thus they can be stable;

[3] The formation of Cooper pairs by trions is outside the scope of the present paper.

[4] One can also consider a triplet state composed of three electrons with parallel spins, with the central one having an antisymmetric envelope function. In our case the quantum well is thin enough to make the energy of a triplet state much higher than the singlet-state energy.

- exciton-polaritons can Bose-condense at very high temperatures because of their light effective mass;
- in systems where the formation of quatron-polariton is possible, the exciton-polariton condensate would relax to the lowest-energy bosonic state, which is a quatron state;
- the stability of quatron-polariton superfluids is dependent on the number of quasi-particles composing the superfluid—if this number reaches a few hundred, stability at room temperature can be assured;
- superfluidity of quatron-polaritons is equivalent to superconductivity.

Acknowledgments

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