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Collective exciton magnetic polarons in quantum wells with semimagnetic barriers

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A mechanism for exciton Bose condensation is suggested. We show that the interaction of localized exciton magnetic polarons (EMP's) can lead to the formation of a free collective exciton magnetic polaron (CEMP) able to move as a whole particle. Critical conditions for this transition and characteristics of the CEMP are found. We interpret the recent data on giant magnetic polaron mobility in $ZnSe/ZnMn_xSe_{1-x}$ quantum wells [G. A. Balchin, C. D. Poweleit, and L. M. Smith, in *Proceedings of the 23rd International Conference on the Physics of Semiconductors*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore 1996), p. 2055] as a manifestation of the free CEMP formation due to clusterization of long-living indirect EMP's. [S0163-1829(98)52308-9]

Exciton magnetic polarons (EMP's) are quasiparticles that are found in crystals having magnetic ions. They are formed due to the short-range coupling between the exciton spin and the spins of these magnetic ions.[1] In recent years, localized EMP's have been found experimentally in quantum wells with semimagnetic barriers like $CdTe/Cd_{1-x}Mn_xTe$ or $ZnSe/ZnMn_xSe_{1-x}$.[2–4] Theory of the EMP's (Refs. 5–7) has demonstrated that, in real structures, a primary nonmagnetic localization of an exciton is necessary for the EMP to be formed. A critical temperature of the free magnetic polaron (FMP) formation appeared to be too low to give any chance of observing FMP's experimentally. It is also known that an attracting force between EMP's exists, due to their interaction with the magnetic media, allowing, in principle, the formation of magnetic bipolarons.[8] The lower the dimensionality of the system, the greater the chance to create a magnetic bipolaron. Until now, no experimental evidence for the existence of magnetic bipolarons or collective polarons has been reported, to the best of our knowledge.

In this paper, a collective EMP formed by an ensemble of N excitons in a condensed phase in a quantum well (QW) with semimagnetic barriers is theoretically considered, to the best of our knowledge for the first time. We show that the critical temperature of the free polaron formation increases proportionally to N, which makes possible the formation of a two-dimensional (2D) collective FMP. In other words, the exchange interaction between excitons and magnetic ions can lead to the exciton Bose condensation at temperatures much higher than the temperature of FMP formation.

The phenomenon of Bose condensation of excitons in quantum wells has been a subject of many works (see, for instance, Refs. 9–11 and the references therein). Here, we only consider the features of the condensed exciton state specific for the case of a collective EMP, and do not discuss the problems of exciton condensation in general. It is worthwhile to mention, however, the reasons why the semimagnetic QW's seem favorable for exciton condensation. A substantial lifetime of excitons is needed to form the condensate, which may be realized in type-II quantum wells where electron and hole are spatially separated. On the other hand, in type-II wells the Coulomb interaction between electrons and holes is weaker, which normally complicates the formation of the condensate.[11] In quantum wells with semimagnetic barriers, the following scenario can be realized. Type-I direct excitons are excited. During the polaron formation the holes transfer into one of the semimagnetic barriers and the excitons become spatially indirect and, consequently, long living. The attraction of indirect excitons does not have a Coulombic nature: It is due to the exchange interaction between excitons and magnetic ions, which is stronger for spatially indirect than for direct excitons, in many cases. In the following, we will discuss whether this mechanism of condensation can be realized in real systems.

Recently, Balchin, Poweleit, and Smith[12] reported an experimental observation of a giant mobility of EMP's in $ZnSe/ZnMn_xSe_{1-x}$ QW's. This seems to be a paradox since localized EMP's (with a binding energy of about 30 meV) should obviously have a negligibly small mobility.[13] Only free magnetic polarons can move in a QW plane. Simple estimations show that the FMP's cannot be formed in the studied system. Our analysis allows us to interpret the experiments[12] as a manifestation of the exchange induced exciton Bose condensation in $ZnSe/ZnMn_xSe_{1-x}$ quantum wells.

The Schrödinger equation for a single $e1hh1$ EMP in a QW with semimagnetic barriers is written as

$$\{K+V+U_{\mathrm{Coul}}+U_{\mathrm{exch}}(\Psi_{\mathrm{exc}})-E\}\Psi_{\mathrm{exc}}=0, \tag{1}$$

where K is the kinetic-energy operator describing electron and hole motion normal to the QW plane direction, their relative motion, and the exciton center of mass motion, V is the QW potential, U_{Coul} is the operator for electron-hole Coulomb interaction, U_{exch} is the wave-function-dependent exchange term, which can be represented as[1]

$$U_{\mathrm{exch}}=(\alpha I-\tfrac{1}{3}\beta J)N_0\tilde{x}\langle s(\Psi_{\mathrm{exc}}^2)\rangle,$$

$$\langle s(\Psi_{\mathrm{exc}}^2)\rangle=\tfrac{5}{2}B_{5/2}\left(\frac{2(\alpha I-\frac{1}{3}\beta J)\Psi_{\mathrm{exc}}^2}{5k(T+T_0)}\right). \tag{2}$$

Here α and β are the electron and hole exchange constants defined in the semimagnetic material (barrier, in our case), I

and J are electron- and heavy-hole spins, $B_{5/2}$ is the Brillouin function, $N_0\tilde{x}$ is an effective concentration of the magnetic ions taking part in the exchange interaction, and T_0 is an effective temperature. Two latter quantities are introduced to take into account the spin-glass effect.[14] k is the Boltzmann constant.

The critical temperature T_c of a free magnetic polaron formation is a characteristic of equilibrium when the loss of the free energy of the system due to exchange interaction is equal to the increase of the exciton kinetic energy if one starts to localize a free exciton. It can be found from condition

$$\left.\frac{\partial F}{\partial(1/R)}\right|_{R\to\infty}=0, \tag{3}$$

where R is the radius of the polaron, and F is the free energy of the system that coincides with the energy, except the exchange term, which has the following form:

$$\tilde{F}_{\rm EMP}=-kTN_0\tilde{x}\int dr \times\ln\left[\frac{1}{6}\sum_{m=-5/2}^{5/2}\exp\left(\frac{m(\alpha I-\frac{1}{3}\beta J)|\Psi_{\rm exc}^2|}{k(T+T_0)}\right)\right]. \tag{4}$$

It has been shown in Refs. 5–7 that

$$T_c+T_0=\eta\,[(\alpha I-\tfrac{1}{3}\beta J)^2]/\hbar^2/M_{\rm exc}, \tag{5}$$

where η is a coefficient dependent on the QW shape, and $M_{\rm exc}$ is the exciton mass.

Suppose that N excitons, all having the same wave function $\Psi_{\rm exc}$, have formed a magnetic polaron. The ground state of this collective polaron can be found from Eq. (1) with a substitution $\Psi_{\rm exc}\to\sqrt{N}\Psi_{\rm exc}$. Here we only consider those excitons that have formed a condensed state, while evidently, at a finite temperature, there are others with nonzero in-plane wave vectors that do not participate in the polaron. Here and further on, we will neglect the correction to the Coulomb energy due to the interaction between excitons supposing that it is small in comparison with the exchange potential variation.[8] Moreover, we fully adopt the approach of Keldysh and Kopaev,[9] who considered spatially indirect excitons as bosons, neglecting the fermionic nature of single electrons and holes. This seems to be valid at concentrations below the Mott transition, i.e., in the excitonic regime, which is likely to be fulfilled at the conditions of the experiment.[12] The exchange contribution to the exciton localization energy is governed by a polarization degree of magnetic ions. The higher the probability of finding an exciton at each magnetic ion, the stronger the magnetic ions are polarized. The probability of finding an exciton obviously increases if instead of one particle, N ones, in the same state, have condensed.

This has an important consequence for the critical conditions of the FMP formation. If we have N excitons instead of only one, the kinetic energy increases proportionally to N, but the exchange energy increases more strongly (proportionally to N^2 in case of a quasidelocalized exciton). Substituting, in Eq. (5), I by NI, J by NJ, and multiplying the kinetic-energy term in the denominator by N, one finds that *the critical temperature of a free magnetic polaron formation increases proportionally to* N.

Until now, we have only discussed the static properties of collective polarons. In order to conclude whether their formation is possible in real systems, one should have some idea of the dynamics of their formation.

The possibility of creating a collective state requires having a few single localized polarons not far from each other and with a long enough lifetime. In the following, we will try to determine more precisely what is ''not far'' and ''long enough'' using basic thermodynamics arguments. Suppose that the quantity of localized polarons in the plane is substantial, $N\to\infty$, and that they have an in-plane concentration n. The formation of the collective polaron is preferential in this case if the free energy of a large-radius polaron (for which the probability of finding an exciton in a definite point in the QW plane equals n) exceeds the free energy of N single polarons. A critical condition for n, therefore, can be written

$$K-U+\tilde{F}_{\rm EMP}=\tilde{F}_{\rm CEMP}, \tag{6}$$

where K and U are the kinetic and potential energies of the exciton center-of-mass wave function in the plane of the QW for a single polaron. The exchange part of the free energy of the collective polaron is

$$\tilde{F}_{\rm CEMP}=-kT\frac{N_0\tilde{x}}{n}\int dz\,\ln\left[\frac{1}{6}\sum_{m=-5/2}^{5/2}\exp\left(\frac{m(\alpha I-\frac{1}{3}\beta J)|U_{\rm exc}^2(z)|n}{k(T+T_0)}\right)\right]. \tag{7}$$

Here $U_{\rm exc}(z)$ is the z component of the exciton wave function.

$\tilde{F}_{\rm CEMP}$ is negative and decreases linearly with the increase of n at small n, then saturates. Thus, in order to make CEMP formation favorable, one should increase the in-plane concentration of the magnetic polarons. The latter depends mostly on two factors: the intensity of excitation G and the exciton lifetime τ: $n\propto G\tau$. Increasing G causes a heating of the system that is unfavorable for the magnetic polaron formation. In order to facilitate the condensation of magnetic polarons, one should try to increase the exciton lifetime. The QW's with semimagnetic barriers offer a great advantage in this sense: In these structures an initially symmetric spatially direct exciton state can spontaneously lose the mirror symmetry during the magnetic polaron formation, so that a type-II spatially indirect exciton is formed that obviously has a much longer radiative lifetime. This effect has been predicted theoretically[15,16] and observed experimentally in $ZnSe/ZnMn_xSe_{1-x}$ QW's.[17] The spontaneous transfer of the heavy hole into one of the semimagnetic barriers takes place at temperatures lower than a critical one that can be found from condition

$$\left.\frac{\partial^2 F}{\partial z_0^2}\right|_{z_0=0}=0, \tag{8}$$

where F is the free energy of the magnetic polaron calculated over all centrosymmetric trial functions, and z_0 is a shift of the hole center of mass from the center of the QW towards a barrier. The critical temperature given by Eq. (8) appears to also be a critical temperature for the collective EMP formation since a spatially direct state has most probably a too short lifetime to allow for the condensation of polarons.

A simple approach of estimating the mobility of a free 3D magnetic polaron has been developed in Ref. 18. The idea was to equalize the work per unit time for the orientation of the spins of magnetic ions made by a moving polaron, with the product of the external force acting on the polaron and its velocity. Note that this model ignores all contributions to the mobility that are not specific for magnetic polarons, i.e., interaction with phonons, scattering at potential fluctuations, etc. Generalizing this approach for a 2D case, one can estimate the magnetic polaron effective mobility as

$$\mu \propto (\mu_B^2 g^2 L_z a^4 e)/\beta^2 \chi \tau, \qquad (9)$$

where μ_B is the Bohr magneton, g is the magnetic ion g factor, L_z is the QW width, a is the radius of the magnetic polaron in a QW plane, χ is the magnetic susceptibility of the semimagnetic semiconductor, and τ is the longitudinal spin-relaxation time of a magnetic ion. For a collective polaron, formula (9) is applicable provided that the external force is uniform over the area occupied by a polaron. For large N the magnetic polaron radius a increases as $\sqrt{N}$ so that an effective mobility is expected to increase as N^2. The hole mobility measured in p-ZnSe is about 25 $\mathrm{cm^2/V\,s}$,[12] thus the effective mobility of a single exciton magnetic polaron is of the order of 10 $\mathrm{cm^2/V\,s}$ (this number is of course not exact because the single polarons are essentially localized and their hopping mobility strongly depends on the scale of magnetic fluctuations in the barriers). Consequently, the effective mobility of the collective polaron formed by 100 excitons is about 100 000 $\mathrm{cm^2/V\,s}$, which is the same order of magnitude as the data of Ref. 12.

In optical experiments with spatial resolution, high polaronic mobility can be detected in the case of efficient diffusion mechanism. EMP's are neutral. Consequently, to make them move, one should create some nonelectric force. Such a force can originate from the repulsion of magnetic polarons with opposite magnetic moments. The mechanism of this repulsion can be described as follows: two excitons with opposite spins separated with some distance R try to orient the magnetic ion spins in the environment in order to form a polaron and to increase their energy, while each exciton orients the ion spins parallel to its own spin. Those ions that are between two polarons remain unpolarized or weakly polarized. It is therefore energetically preferential for the excitons to move away from each other in order to be able to orient all magnetic ions within their localization areas. In Ref. 8, the attraction of two magnetic polarons having parallel spins has been analyzed. Clearly, the repulsive potential in the case of opposite spins has the same shape but the opposite sign. Thus, one can derive the repulsive force between two identical collective polarons P as the function of the distance between their centers R in the form

$$P(R) = \frac{TN_0\widetilde{x}}{3(T+T_0)}\left(\alpha I - \tfrac{1}{3}\beta J\right)\int d^3r\, \frac{\partial |\Psi_{\mathrm{exc}}(r-R)|^2}{\partial R} \times \langle s[N|\Psi_{\mathrm{exc}}(r)|^2 - N|\Psi_{\mathrm{exc}}(r-R)|^2]\rangle . \qquad (10)$$

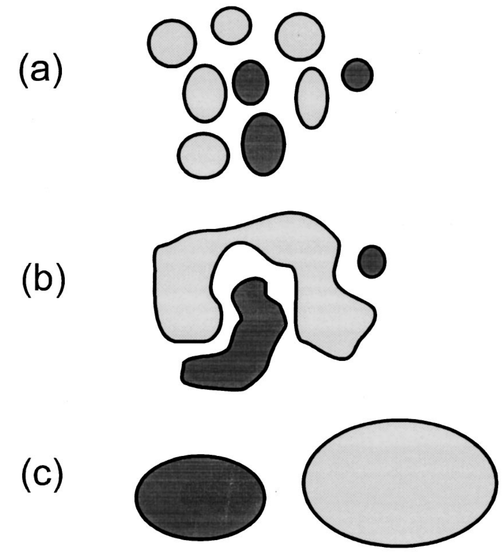

FIG. 1. The schema of the collective EMP formation in a QW with semimagnetic barriers: (a) localized single EMP's are forming, (b) the polarons with parallel magnetic momenta clusterize, and (c) repulsion between clusters makes them move away from each other. EMP's with opposite directions of magnetic momentum are distinguished by using light and dark gray.

Note that the force $P(R)$ is normalized to a single exciton. The factor of N (number of excitons in the collective polaron) enters in the average magnetic ion spin $\langle s \rangle$, which is defined in the same way as in Eq. (2). One can see that the repulsive interaction of polarons with opposite spins becomes stronger with the increase of N, which is quite understandable since the radius of polarons increases with N.

In reality, however, in the case of a nonpolarized excitation, excitons having opposite spins are randomly distributed in the QW plane, so that the formation of collective polarons and their repulsion represent a more complicated process as shown schematically in Fig. 1. First, the single localized polarons are forming. Then they organize clusters of free polarons. Then the repulsion between clusters leads to the formation of more or less cylindrically symmetrical polaronic drops.

In order to get a numerical idea about the characteristics of the collective polaron state, we have performed for the structure for which Balchin, Poweleit, and Smith[12] have found a surprising giant mobility of magnetic polarons, i.e., a $\mathrm{ZnSe/Zn_{0.86}Mn_{0.14}Se}$ QW of 10 nm thickness, a self-consistent variational calculation using a modified version of the technique described in Ref. 19.

We have solved the Schrödinger equation (1) for a collective polaron substituting $\Psi_{\mathrm{exc}} \rightarrow \sqrt{N}\Psi_{\mathrm{exc}}$ in the exchange term (2) and taking account only of the hole contribution since the exchange constant for an electron is much smaller than for a hole. The self-consistent Hamiltonian (1) has been applied to a trial function

$$Y_{\mathrm{exc}} = U_e(z_e) U_h(z_h) F_\gamma(R) \Phi_\lambda(\rho, z_e - z_h), \qquad (11)$$

where $U_e(z_e)$ and $U_h(z_h)$ are electron and hole envelope functions normal to the QW plane direction,

$$F_\gamma(R) = \sqrt{(2/\pi\gamma^2)}\, \exp(-R^2/\gamma^2)$$

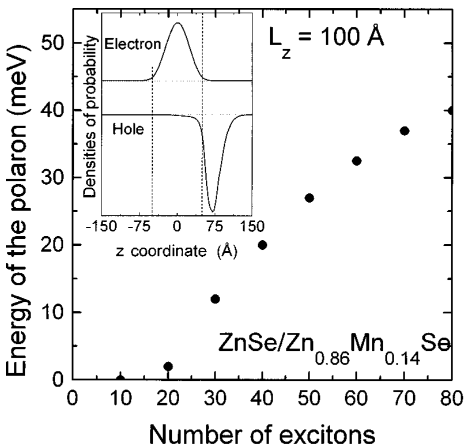

FIG. 2. The energy of the free collective EMP formed in the $ZnSe/Zn_{0.86}Mn_{0.14}Se$ QW as a function of the number N of excitons taking part in the polaron formation. The inset shows the QW potential and electron- and heavy-hole envelope functions normal to the QW plane direction for $N=50$.

is the exciton center of mass wave function in a QW plane, and

$$\Phi_\lambda(\rho,z_e-z_h)=\exp\{-\sqrt{\lambda[\rho^2+(z_e-z_h)^2]}\}$$

describes the electron-hole relative motion. γ and λ are the variational parameters that are obtained self-consistently and represent physical probes of the polaron in-plane extension and of the exciton binding energy.

The parameters used are electron- and heavy-hole band offsets $V_e=45$ meV and $V_h=20$ meV, $T_0=4.4$ K, $\tilde{x}=0.08$, $\alpha N_0=0.26$ eV, $\beta N_0=-1.34$ eV, the electron mass $m_e=0.16m_0$, the heavy-hole mass $m_{hh}=0.49m_0$, the light-hole mass $m_{lh}=0.145m_0$, $L_z=10$ nm, and $T=2$ K. Figure 2 shows the free collective EMP energy as function of number of excitons participating in its formation N. One can see that for N less than 15, the free polaron cannot be formed while for greater N its energy increases with the increase of N saturating at the value $E_{CEMP}=45$ meV. This is in agreement with the calculation of the critical temperature of the FMP formation by formula (5) (see also Ref. 7). For a single FMP the critical temperature appeared to be $T_c+T_0=0.4$ K. Since this quantity increases linearly with N, as it has been shown above, at helium temperature the free polaron can be formed by $N>15$ excitons. The inset in Fig. 2 shows the calculated electron and hole envelope functions in normal to the QW plane direction for $N=50$. One can see that the exciton is strongly spatially indirect, which provides its sufficiently long lifetime (in the order of 10 ns for the given conditions of localization). According to Eq. (9), the critical temperature of the spatially indirect EMP formation is estimated to be about 10 K, which is very close to the temperature at which the EMP mobility experiences a steplike decrease according to the data.[12] The critical in-plane concentration of localized single EMP's necessary for the collective polaron formation is estimated from Eq. (6) as $n\approx 2.10^{-3}$ nm^{-2}, with a single localized polaron free energy taken to be 10 meV, which means that the average distance between localized excitons should be approximately three times more than the exciton Bohr radius. This condition is likely to be realized in a QW excited by a laser pulse if the exciton lifetime is of the order of tens of ns.

Very recently, an anomalous negative magnetoresistance in (Zn,Cd)Se/ZnSe quantum wells doped with manganese has been reported[20] and associated with unbinding of *free* magnetic polarons in a two-dimensional electron gas. Moreover, a strong photoinduced increase of magnetization and conductivity at some critical concentration of holes has been found in a new semimagnetic semiconductor heterosystem (In,Mn)As/GaSb.[21] This may be two new experimental confirmations of the stability of collective free magnetic polarons in low-dimensional systems.

In conclusion, a Bose-condensed exciton state can be formed due to the interaction between localized excitons via their exchange coupling with magnetic ions in a QW with semimagnetic barriers. The necessary conditions for this effect are (i) a long enough exciton lifetime, which is realized at low temperatures because of the exchange induced transfer of the hole into one of the semimagnetic barriers, and (ii) substantial concentration of the excitons in the QW, which also requires their long lifetime. These conditions are likely to be realized in the $ZnSe/Zn_{0.86}Mn_{0.14}Se$ heterosystem recently studied.[12] Our variational calculation allows interpreting their data on a giant mobility of magnetic polarons in terms of the collective EMP formation.

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