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THE EXCITONIC SPECTRUM OF GERMANIUM IN A STRONG MAGNETIC FIELD

Yu.P.Kravchenko, M.A.Liberman

Department of Physics, Uppsala University
S-751 21, Uppsala, Sweden

P.Kapitsa Institute for Physical Problems, RAS
117334 Moscow, Russia

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As was found in recently reported experiments with the uniaxially deformed germanium in a magnetic field [7], sufficiently strong intensity of the applied magnetic field results in the appearance of a new line in the optical spectrum of the excitons. We propose a mechanism which can provide explanation of this experimentally observed spectral feature. The new spectral line may be attributed to the formation of strongly bound biexcitonic molecules in the quantum state $^3\Pi_u$.

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Studies of the behavior of matter in strong magnetic fields constitute one of the most interesting and rapidly developing areas in atomic and molecular physics [1-4]. The interest in this area is motivated by those radical changes in the electronic structure and properties of matter which occur when the paramagnetic energies of electrons become comparable to the typical energies of atomic and molecular bonds or exceed those energies. In the case of usual atoms and molecules corresponding magnetic fields belong to the astrophysical domain. Indeed, the typical atomic binding energy may be estimated as 1 hartree $= m_e e^4/\hbar^2 = 27.2$ eV, while the paramagnetic energy of the electron is the distance between Landau levels, equal to $\hbar\omega_H = \hbar eH/m_e c$, where ω_H is the cyclotronic frequency and H is the magnetic field intensity (we shall use CGC units). By equating these two expressions, we immediately find that the critical intensity of "strong" magnetic field is $H_0 = m_e^2 e^3 c/\hbar^3 = 2.35052 \cdot 10^9$ G. Being far beyond reach by laboratory standards, magnetic fields of such intensity are, nevertheless, quite usual on the astrophysical scale. Magnetic fields in the vicinity of magnetic stars reach 0.1 H_0, while magnetic fields on the surfaces of neutron stars and pulsars can exceed H_0 by three orders of magnitude. For convenience, we shall measure the magnetic field in atomic units as $\gamma = H/H_0$.

This scale of magnetic fields profoundly changes, however, if one turns to the behavior of hydrogen-like excitons in semiconductors. First, the electron mass m_e changes to the reduced mass of electron-hole pair $m = m_e m_h/(m_e + m_h)$. Second, the binding energy of excitons is reduced by ε^2 times, where ε is the dielectric constant of the semiconductor. As a consequence, the critical intensity of the magnetic field is reduced to $H_0^* = m^2 e^3 c/\varepsilon^2 \hbar^3$. This quantity is about several tesla for Ge and Si and may be as small as 0.2 T for InSb. Such a low value of the critical field implies that the structure of excitons in semiconductors must experience significant changes already in laboratory magnetic fields.

Such changes have indeed been observed in many laboratory experiments. One line of the experimental enquiry concentrates on the excited levels of excitons in semiconductors with relatively high values of the critical magnetic field. A typical such a material is Cu_2O

with critical magnetic field $H_0^* \sim 800$ T, which demonstrates the "chaotic" behavior of excited excitonic levels in laboratory magnetic fields of the order $10^{-2}H_0^*$ [5]. Another line of research is the studies of excitons in semiconductors with critical magnetic fields comparable to or lower than the magnetic fields attainable in the laboratory. A representative example is the uniaxially deformed germanium, which is especially interesting due to the fact that this material allows to investigate the influence of the magnetic field not only on separate excitons but on excitonic molecules as well [6].

Recent experiments with Ge in magnetic fields up to 14 T revealed new interesting features of the excitonic spectrum [7]. The authors of [7] studied the optical spectra of excitons in the uniaxially deformed germanium, which was placed in a magnetic field. The critical magnetic field intensity for Ge was $H_0^* = 2.9$ T. In the absence of magnetic field, the excitonic spectrum consisted of two lines, the line of excitons and the accompanying line of biexcitonic molecules. Application of the magnetic field resulted in the decrease of intensity of the biexcitonic line and its final disappearance at ≈ 1.5 T, which corresponds to the effective magnetic field $\gamma \approx 0.5$ a.u. When the applied magnetic field reached 4 T ($\gamma \approx 1.4$ a.u.), a new spectral line has appeared. It was located on the "red" side of the line of free excitons and was labeled as the "X-line". This line was associated with the appearance of another bound state, whose energy is by one electron-hole pair lower than the energy of an isolated exciton.

The authors of [7] proposed two possible mechanisms explaining the observed spectrum. The first one was based on the assumption of the increased stability of electron-hole liquid in magnetic field, which they doubted. An alternative explanation was the formation of a new biexcitonic molecular state. Although this possibility seemed promising, further progress in that direction was hindered by the lack of information about the behavior of excitonic (and hydrogen) molecules in strong magnetic fields. Indeed, even such a basic question as the symmetry of the ground state of H_2 in magnetic field was subjected to a prolonged dispute. However, recent calculations of the electronic structure of the hydrogen molecule in strong magnetic fields reveal more detailed information about the electronic states of H_2 and allow us to offer a possible explanation of the nature of the new spectral line, described in [7].

We suggest that the explanation of the observed phenomenon is the appearance of metastable excitons in the quantum state $^3\Pi_u$. In what follows, we shall consider the electronic structure of the hydrogen molecule and use the fact that the hydrogen data can be scaled to describe the behavior of hydrogenlike excitons. The analysis is based on the Hartree–Fock calculations reported in [8]. The molecular axis is directed along the magnetic field and the nuclei are assumed to be infinitely heavy. It was found in [8] that, when the magnetic field increases, the ground state experiences two symmetry transitions. The first transition occurs at $\gamma \approx 0.18$ a.u., when the ground state changes from the strongly bound singlet state $^1\Sigma_g^+$ to the weakly interacting triplet state $^3\Sigma_u^+$. The second transition happens at $\gamma \approx 14$ a.u. and involves the change from $^3\Sigma_u^+$ to the strongly bound triplet state $^3\Pi_u$. What is important for the present analysis, however, is the fact that, starting from $\gamma \approx 1.2 - 1.4$ a.u., the strongly bound state $^3\Pi_u$ may be the *metastable* state of the molecule. Therefore, starting from these intensities of the effective magnetic field, one can expect formation of metastable biexcitons, which, as we believe, provide explanation of the "X-line", observed by Timofeev and Chernenko.

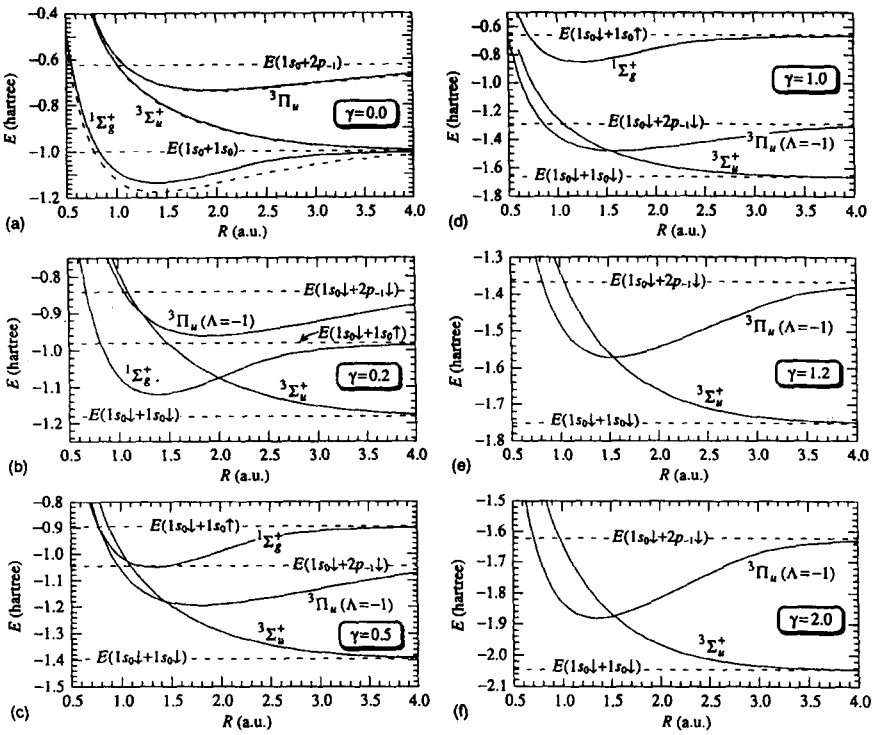

Fig.1 Potential curves of the electronic states $^1\Sigma_g^+$, $^3\Sigma_u^+$, and $^3\Pi_u$ ($\Lambda = -1$) of the hydrogen molecule in a strong parallel magnetic field γ: (a) $\gamma = 0.0$, (b) $\gamma = 0.2$, (c) $\gamma = 0.5$, (d) $\gamma = 1.0$, (e) $\gamma = 1.2$, and (f) $\gamma = 2.0$ a.u. Dotted lines show energies of the corresponding states at infinite nuclear separation. For the triplet states $^3\Sigma_u^+$ and $^3\Pi_u$, the total electronic spin is antiparallel to the magnetic field, two other components of the triplet are not shown

Let us consider how these ground state transitions occur. Fig.1*a* shows the potential curves $^1\Sigma_g^+$, $^3\Sigma_u^+$, and $^3\Pi_u$ of the hydrogen molecule in the absence of magnetic field. The internuclear distance R is measured in the units of the Bohr radius $a_0 = \hbar^2/m_e e^2 = 5.3 \cdot 10^{-9}$ cm, and the zero of energy corresponds to the situation where all particles are at the infinite separation from each other in their lowest Landau states with their spins antiparallel to the magnetic field. Dashed curves show the total energy with account for the electron correlation, solid curves correspond to the Hartree–Fock approximation. While the correlation energy is significant for the singlet state $^1\Sigma_g^+$, its value for triplet states is much smaller due to the smaller overlap of electronic orbitals. It should be noted that the correlation energy remains approximately the same even in the presence of magnetic fields up to $\gamma \lesssim 1$ a.u. This can be confirmed by comparing our Hartree–Fock results with detailed configuration-interaction calculations of Σ-states of H_2 in magnetic field, which were recently reported in [9].

Fig.1*b* shows the potential curves of the same three quantum states in the presence of the parallel magnetic field $\gamma = 0.2$ a.u. We see that the symmetry of the ground state has changed. The singlet state $^1\Sigma_g^+$, which is the ground state at $\gamma = 0$, has been slightly shifted up, while the weakly interacting triplet state $^3\Sigma_u^+$ has been shifted lower due to the increase in the binding energy of isolated hydrogen atoms. As a result, the minimum

of the potential curve of $^1\Sigma_g^+$ now lies *above* the energy of $^3\Sigma_u^+$ at $R \gg 1$, and the latter state represents the true ground state of the system. However, the hydrogen still can form tightly bound molecules in the state $^1\Sigma_g^+$, but such molecules will be metastable. As for the state $^3\Pi_u$, it lies above the potential curves of both $^1\Sigma_g^+$ and $^3\Sigma_u^+$ and is therefore unstable.

If we now increase the field to $\gamma = 0.5$ a.u., we encounter further changes. Fig.1*c* shows that the potential curve of the singlet state $^1\Sigma_g^+$ has shifted even higher, and $^1\Sigma_g^+$ is now an unstable state. This is manifested by the disappearance of biexcitonic line at $\gamma \approx 0.5$ a.u., observed in [7]. The potential curve of the state $^3\Pi_u$ is still above that of the state $^3\Sigma_u^+$. The hydrogen cannot form strongly bound molecules, and the ground state $^3\Sigma_u^+$ of the molecule is represented by two separated atoms. The molecule is bound very weakly, if at all, and the hydrogen now presents a gas of weakly interacting atoms, which may exhibit such phenomena as Bose condensation and superfluidity [2].

This situation remains essentially the same in the magnetic field $\gamma = 1$ a.u., as shown on Fig.1*d*. However, the potential well of the triplet state $^3\Pi_u$ has deepened, and its minimum now lies only slightly above the curve of the state $^3\Sigma_u^+$. When we increase the magnetic field to $\gamma = 1.2$ a.u., the potential minimum of $^3\Pi_u$ crosses the energy curve of $^3\Sigma_u^+$ (Fig.1*d*). This means that if the field increases even further, the hydrogen can start to form strongly bound metastable molecules in the state $^3\Pi_u$. Fig.1*f* shows that in the magnetic field $\gamma = 2.0$ a.u. the potential minimum of $^3\Pi_u$ lies below the potential curve of the weakly interacting state $^3\Sigma_u^+$, which is a typical picture of a metastable state.

Therefore, the quantum state $^3\Pi_u$ is an excellent candidate for the biexcitonic state, which can be responsible for the appearance of the "X line" observed in [7]. The most convincing argument in the support of this claim is that the strength of the magnetic field at which the "X-line" was first observed (4 T) corresponds to the effective magnetic field $\gamma \approx 1.4$ a.u. Presented calculations show that the state $^3\Pi_u$ becomes metastable starting from $\gamma \gtrsim 1.2$ a.u. It is unlikely that such a perfect agreement between the two values is occasional. However, in order to verify this claim one needs to perform similar experiments with other semiconductors having different values of the critical magnetic field.

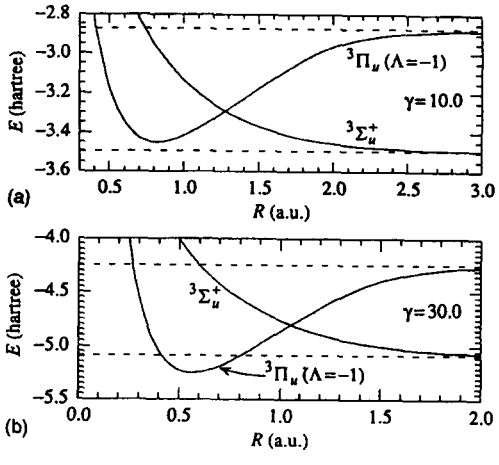

Fig.2. Potential curves of the triplet states $^3\Sigma_u^+$and $^3\Pi_u$ ($\Lambda = -1$) of the hydrogen molecule in the magnetic field: (a) $\gamma = 10.0$ a.u., (b) $\gamma = 30.0$ a.u. All designations are as on Fig.1 .

Let us follow the further evolution of the states $^3\Sigma_u^+$ and $^3\Pi_u$. Fig.2 shows their potential curves in the magnetic field $\gamma = 10$ a.u. and $\gamma = 30$ a.u. We see that at $\gamma = 10$ a.u. the true ground state of the molecule is still the weakly interacting state $^3\Sigma_u^+$. At $\gamma = 30$ a.u. the potential minimum of the state $^3\Pi_u$ lies below the minimum of $^3\Sigma_u^+$, and the ground state of the system is the strongly bound $^3\Pi_u$. Calculations show that this second transition of the ground state symmetry occurs at $\gamma \approx 14$ a.u. [8].

Let us summarize our conclusions. We have proposed a possible theoretical explanation of the nature of the new excitonic line, observed experimentally in [7]. According to our calculations, this new "X-line" can be explained by the formation of metastable biexcitonic molecules in the quantum state $^3\Pi_u$. The calculated strength of the magnetic field, in which such metastable molecules can exist, is an excellent agreement with the experimental results of [7]. If the proposed mechanism is correct, it should work for other semiconductors with hydrogen-like excitons and manifest itself at the same effective magnetic field strength $\gamma \approx 1.4$ a.u. If such an effect will be observed in experiments with other types of semiconductors, it may open alternative possibilities for the control of the optical spectrum of semiconductors by the applied magnetic field. Since certain kinds of semiconductors have very low values of the critical magnetic field, this mechanism may have potential technological applications.

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