

Megagauss magnetospectroscopy of EuS/PbS multi-quantum wells

I. Stolpe, N. Puhmann, O. Portugall, and M. von Ortenberg

Institute of Physics of the Humboldt University at Berlin, Invalidenstrasse 110, D-10115 Berlin, Germany

W. Dobrowolski

Institute of Physics, Polish Academy of Sciences, Aleja Lotników 32/46, 02-668 Warsaw, Poland

A. Yu. Sipatov

State Technical University of Kharkov, Frunze 21, 310002 Kharkov, Ukraine

V. K. Dugaev

Chernovtsy Branch of the Institute of Materials Science Problems, Ukrainian Academy of Sciences, Vilde 5, 58001 Chernovtsy, Ukraine

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Magnetotransmission studies of EuS/PbS multiquantum wells in magnetic fields up to 200 T and in the temperature range from 5 to 300 K are reported. A series of transitions are observed, which we interpret as cyclotron resonance transitions, i.e., the transitions between the lowest magnetic subbands. With this identification, the positions of observed resonances are satisfactorily described by theory when the quantum well width is larger than 100 Å. For narrower quantum wells, however, the discrepancy between theory and experimental measurements is significant. Possible explanations for this discrepancy are discussed.

I. INTRODUCTION

In the past several years multilayer structures comprised of alternating ferromagnetic and diamagnetic layers have attracted considerable attention due to their possible applications as materials for spin electronics.¹ Apart from the giant magnetoresistance observed in metallic multilayer structures, physical effects related to the emission of spin-polarized electrons from a ferromagnet into a diamagnet are anticipated to occur in semiconductor-ferromagnet multilayer structures.²

An important property of a semiconductor layer in such nanostructures, as compared to a metallic layer, is the role of quantization of the electron energy spectrum. This effect is not so important for metals since in a metal the electron wavelength is considerably shorter than in a semiconductor. In the context of spin-polarized electron effects, the spin dependence of such quantized electronic levels provides the basis for understanding the spin injection effects in magnetic/nonmagnetic structures. In particular, we believe that the ferromagnetic coupling between magnetic layers separated by a thin nonmagnetic semiconductor layer can also be understood by taking into account the quantization of levels in a semiconducting quantum well (QW).³

Here we present the results of an investigation of EuS/PbS multilayers by high field magnetospectroscopy (HFMS). It has recently been shown^{4,5} that such structures can be considered as a model low-dimensional Heisenberg ferromagnetic system. EuS is a large-gap magnetic semiconductor ($E_g=1.6$ eV), which orders ferromagnetically at $T_c=16.5$ K. PbS is a narrow gap ($E_g=0.3$ eV) degenerated semiconductor. Both EuS and PbS grow in rocksalt structure with nearly perfect lattice matching ($\Delta a/a=0.6\%$). In terms of the electronic potential profile, in this system the PbS

layers correspond to quantum wells, and the EuS layers to the barriers.

Electrons within the PbS QW's have a discrete energy spectrum due to the quantization of motion perpendicular to the layer planes. This size-quantization effect is especially pronounced in narrow gap semiconductors (such as PbS) due to the small value of the effective mass of the electrons. However, the mathematical description of such wells is rather complex, because the IV-VI narrow gap semiconductors are characterized by both a strong nonparabolicity and a strong spin-orbit interaction.⁶

In earlier studies, the energy spectrum of a PbS QW in the PbS/EuS system has been studied by photoluminescence.⁷ Here we explore another powerful method of investigating this system—high field magnetospectroscopy. As has been shown in Refs. 8 and 9, this method provides a powerful additional tool for studying size quantization. In particular, using magnetic fields in the megagauss range ($B>100$ T) makes it possible to study size-quantization effects in systems comprised of layer thicknesses in the range of 1 to 2 nm,⁸ since these lengths exceed the magnetic length for that field range. An important advantage of this technique is that the megagauss spectroscopy can be carried out in a wide temperature range, whereas luminescence measurements are usually restricted to low temperatures. In particular, HFMS enables one to check the correctness of the theoretical models used to describe the size-quantized energy spectrum of IV-VI-based semiconductor nanostructures in the presence of a magnetic field.¹⁰

II. EXPERIMENT

The epitaxial EuS/PbS multilayers investigated in this work were grown from the vapor phase on BaF₂ substrates on a PbS buffer layer approximately 100 Å thick. EuS was

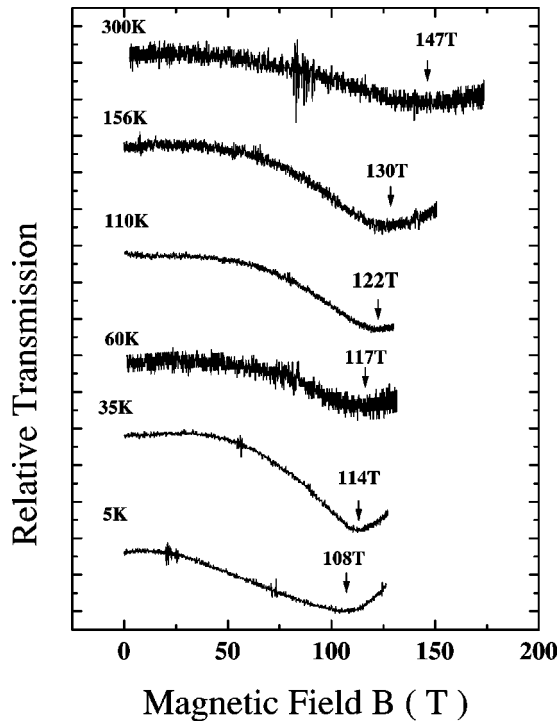


FIG. 1. Transmission spectra for the EuS(100 Å)/PbS(100 Å) multi-quantum well as a function of magnetic field obtained for a series of temperatures. The data were taken at $10.6 \mu\text{m}$ using a CO_2 laser.

evaporated in vacuum (10^{-6} – 10^{-7} Torr) using an electron gun. The growth rate (and thus the layer thickness) were determined *in situ* by a calibrated quartz resonator. The thickness of EuS and PbS monolayers was determined with an accuracy of 1 ML. The quality of the layers and the superlattice period were both checked by x-ray diffraction. The interdiffusion in such types of EuS-PbS structures was also investigated by x-ray diffraction, which allowed us to study composition profile changes as a function of annealing temperature, yielding an intermixed region of roughly 2 ML for unannealed samples.¹¹ All samples are of *n*-type having electron concentrations of the order of 10^{18} cm^{-3} .

The results of magnetic studies of our EuS/PbS structures were reported elsewhere.^{4,5} The magneto-optical experiments described and analyzed in this article were performed in the Berlin single coil megagauss generator¹² in fields up to 200 T. Three structures were investigated: with well widths of 15 Å, 100 Å, and 550 Å, and barrier thicknesses, respectively, of 40 Å, 100 Å, and 700 Å. The samples were mounted in a continuous flow cryostat, which enabled precise control of the sample temperature (T) in the range from 5 to 300 K. Transmission measurements were performed in the Faraday geometry, with B oriented along the (111) direction, perpendicular to the layer plane. A CO_2 laser operating at $10.6 \mu\text{m}$ was used as the radiation source. A fast HgCdTe detector was used for the detection of the transmitted radiation intensity.

Figure 1 shows the transmission as a function of magnetic field B for several temperatures in the EuS(100 Å)/PbS(100 Å) sample. Note that very small changes of transmission (even on the scale of 2%) are clearly resolved.

We attribute the observed resonances to inter-Landau-

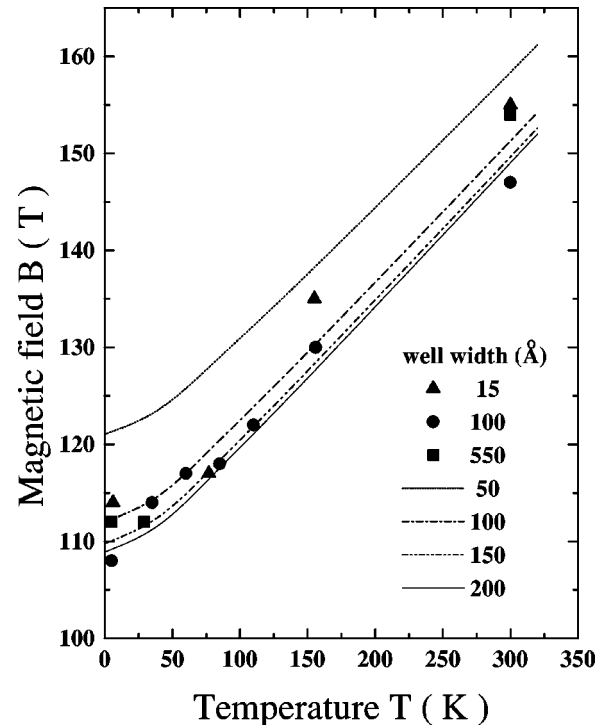


FIG. 2. Magnetic field positions of transmission minima vs temperature for three EuS/PbS multi-quantum wells. The points are experimental. The curves are theoretical, obtained as discussed in the text.

level transitions within the lowest subband in a quantum well, since at such high fields only the lowest Landau level is occupied with electrons. The nonmonotonic dependence of the amplitude of the observed minima on temperature can be explained as an effect of interferences occurring in the substrate of the structure. Figure 2 shows the experimentally observed positions of the resonances observed for several samples, along with theoretical calculations obtained as discussed below.

III. LEVEL QUANTIZATION IN QUANTUM WELLS OF EuS/PbS MULTILAYERS IN PERPENDICULAR MAGNETIC FIELD: THEORY

To calculate the energy levels for the EuS/PbS multi-quantum well system, we use a simple model of a symmetric quantum well, which takes into account both the nonparabolicity of the E vs k dispersion and the spin-orbit interaction of a IV-VI semiconductor.¹⁰ The model neglects a possible anisotropy of the energy spectrum, which is allowed by the crystal symmetry but is very small for PbS compound.⁶

To account for both the size and the magnetic (i.e., Landau) quantization, we use the analytical model developed earlier¹⁰ for the energy spectrum of IV-VI semiconductor quantum wells. In this model the Hamiltonian of the bulk semiconductor has the form of a relativistic Dirac Hamiltonian, the energy spectrum is taken to be isotropic, and the quantum well is modeled by a rectangular potential.

The calculation of the spectrum in a magnetic field perpendicular to the layers is performed in two steps. First, the levels ϵ_n resulting from size quantization are found numerically by solving the equation^{13,14}

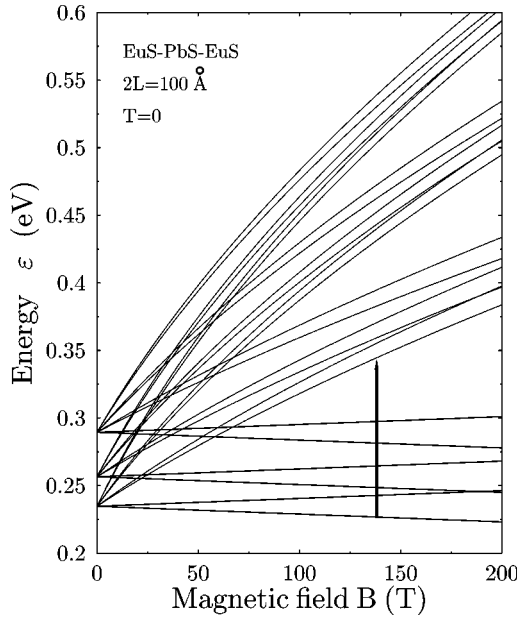


FIG. 3. Results of calculation of the lowest energy levels in a EuS/PbS quantum well as a function of magnetic field applied perpendicular to the layer plane. The arrow indicates the cyclotron-resonance transition originating from the lowest Landau levels.

$$\left(\frac{\kappa_1 \Delta_0 + \epsilon_n}{\kappa_0 \Delta_1 + \epsilon_n} - \tan(\kappa_0 L) \right) \left(\frac{\kappa_0 \Delta_1 + \epsilon_n}{\kappa_1 \Delta_0 + \epsilon_n} + \tan(\kappa_0 L) \right) = 0, \quad (1)$$

where $2\Delta_1$ and $2\Delta_0$ are the energy gaps of bulk EuS and PbS, respectively, $\kappa_0 = (\epsilon_n^2 - \Delta_0^2)^{1/2}/v$, $\kappa_1 = (\Delta_1^2 - \epsilon_n^2)^{1/2}/v$, v is the interband coupling parameter,⁶ and $2L$ is the well width.

Having established the levels ϵ_n which arise from size quantization, the magnetic splitting of each size-quantized level is found using the following expressions¹⁰

$$\epsilon_{nm}^{(\uparrow)} = \frac{g_v - g_c}{4} \mu_B B \pm \left[\left(\epsilon_n - \frac{g^*}{2} \mu_B B \right)^2 + \frac{2mv^2 eB}{\hbar c} \right]^{1/2}, \quad (2a)$$

$$\epsilon_{nm}^{(\downarrow)} = -\frac{g_v - g_c}{4} \mu_B B \pm \left[\left(\epsilon_n + \frac{g^*}{2} \mu_B B \right)^2 + \frac{2mv^2 eB}{\hbar c} \right]^{1/2}, \quad (2b)$$

where $m=0,1,2,\dots$ labels magnetic sublevels of each ϵ_n , g_c and g_v are the g -factors defining the Zeeman splitting of the conduction and valence bands, μ_B is the Bohr magneton, and g^* stands for $(g_c + g_v)/2$. Each size-quantized level gives two sets of Landau levels, $\epsilon_{nm}^{(\uparrow)}$ and $\epsilon_{nm}^{(\downarrow)}$, due to the removal of spin degeneracy by Zeeman splitting.

The calculated dependence of energy levels on magnetic field B is presented in Fig. 3 for the first four spin-up and spin-down Landau levels for the three lowest size-quantization levels. In the calculations we used the following parameters:⁶ $E_{g0} = 2\Delta_0 = 0.331$ eV (PbS, $T=0$), $E_{g1} = 2\Delta_1 = 1.67$ eV (EuS), $v = 3.85 \times 10^{-8}$ eV·cm, and $g_v = g_c = 2$. The value of v is chosen to achieve the best fit to the experimental data.

As seen in Fig. 1, the field at which the cyclotron resonance transition occurs shows a strong temperature depen-

dence. The temperature can enter the effect in several ways. First, the two energy gaps E_{g0} and E_{g1} depend on T . The dependence of E_{g1} on T can be neglected, however, since for $E_{g1} \gg E_{g0}$ the lowest energy levels ϵ_{nm} do not depend significantly on E_{g1} . The dependence of E_{g0} on T for PbS can be included in the calculation in the form⁶

$$E_{g0} = 0.291 + [4 \times 10^{-4} + (6 \times 10^{-4} T)^2]^{1/2} \text{ (eV)}. \quad (3)$$

Second, due to the thermal expansion of the materials, the quantum well width $2L$ will also depend on T . However, this dependence turns out to be too weak to have any measurable effect on the energy levels. We can estimate the change in the well width as¹⁵

$$L = L_0(1 + \alpha T), \quad (4)$$

where $\alpha \approx 1.9 \times 10^{-5} \text{ K}^{-1}$ at 160 K; i.e., the change in well width due to temperature is of the order of at most 5×10^{-3} , which is too small. Thus, the major contribution to the observed shift of the resonance line with temperature comes from the temperature dependence of E_{g0} .

The magnitude of the magnetic splitting is calculated as the difference in energy between the lowest magnetic levels with the same spin, i.e., the cyclotron resonance energy (see arrow in Fig. 3). As noted earlier, the present measurements of the resonance magnetic field as a function of T were carried out for the wavelength $\lambda = 10.6 \text{ } \mu\text{m}$, so that the energy splitting at the observed resonance ΔE is 0.117 eV. The results of the calculations are presented in Fig. 2, along with the experimental points for several samples with different well widths.

IV. DISCUSSION

As can be seen from Fig. 2, the agreement between theoretical curves and experimental points is quite satisfactory for wide QW's ($2L > 100 \text{ } \text{\AA}$). The dependence of E_{g0} on T , as described by Eq. (3), is responsible for the temperature dependence of the resonance frequencies. Other factors give only a small contribution, which can be neglected on the scale of the observed temperature shifts.

It is evident, however, that the observed dependence of the resonance positions on the QW width is weaker than predicted by the model. From theoretical curves it follows that the dependence on the QW width should indeed be weak if $2L > 100 \text{ } \text{\AA}$. However, significant sensitivity to QW width is expected for narrow wells. Here the experimental results observed on the sample with QW width of $15 \text{ } \text{\AA}$ distinctly depart from the theory, since the experimental points lie clearly below the theoretical curve even for $2L = 50 \text{ } \text{\AA}$. A possible explanation of this discrepancy may lie in the fact that in that specific case the barrier thickness is also relatively small ($40 \text{ } \text{\AA}$). Then, due to the interwell interaction (tunneling) of electrons through such thin EuS barriers, each quantum level is broadened into a miniband, thus leading to a smaller value of the energy at the edge of the miniband.

Furthermore, low-temperature behavior of the experimental curves can be affected by strain, which is a source of some nonlinearity. Indeed, each of the PbS/EuS systems studied here has been grown on a BaF₂ substrate,^{4,5} which has a slightly smaller thermal expansion coefficient than

PbS. Thus, when the temperature decreases, a tensile in-plane stress acts on the PbS layer (as described in Ref. 5). Such stress leads to an expansion of the PbS layers in the x - y plane, along with some relatively small compression in the z direction. Such expansion would result in a shift of the size-quantization levels to higher energy values and, in turn, to a decrease of the splitting observed in magnetic field. This can therefore be a source of some deviation of the theoretical curve from a linear dependence at $T < 50$ K.

It should be noted that in our calculations we have used a simplified model, which does not take into account real band offsets for EuS/PbS structures. In accordance with luminescence data,⁷ the QW is nonsymmetrical with respect to the conduction-valence band alignment: the valence-band offset parameter is about 0.1 eV.

Summarizing, we explain the temperature dependence of the resonance magnetic field B by using the dependence of the energy gap E_{g0} on T for PbS. Due to the nonparabolicity of the energy spectrum, the diamagnetic splitting decreases

with increasing size-quantization energy, as is seen from Eqs. (2a) and (2b). Thus, the resonance condition corresponds to a larger magnetic field.

The dependence of the resonance field on the QW width (thickness of the PbS layer) is also related to the nonparabolicity of the energy spectrum: the smaller is L , the larger is the energy of the size quantization, and, in turn, the smaller is the diamagnetic splitting. However, for very thin QW's, the experimental dependence on L turns out to be weaker than predicted by the theory.

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