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Magnetoresistance and Electroluminescence near the Curie–Weiss Temperature in the $Zn_{1-x}Mn_xTe$ Light Emitting Devices

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The light emitting devices based on the p-$Zn_{1-x}Mn_xTe$ bicrystals have been fabricated. The $Zn_{1-x}Mn_xTe$ devices produce red and green emission originating from the internal d-shell transitions in the Mn^{2+} ions and the donor–acceptor pairs recombination, respectively. A critical behavior of the magnetic field dependence of the green emission intensity and a positive magnetoresistance near the Curie–Weiss temperature in the $Zn_{1-x}Mn_xTe$ devices was observed.

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1. Introduction

In this communication we report the fabrication of unipolar light emitting devices (LEDEs) based on the Bridgman grown p-$Zn_{1-x}Mn_xTe$ bicrystals doped with phosphorus (P) to a level of 5×10^{18} cm^{-3} and present results of the electroluminescence (EL) and magneto-EL measurements. We demonstrate that the electrostatic barriers at the $Zn_{1-x}Mn_xTe$ grain boundary (GB) can be used to excite the EL. At temperatures $T \geq 4.2$ K the negative magnetoresistance (MR) in the device is proportional to the Zeeman splitting. However, near the Curie–Weiss temperature the formation of the bound magnetic polarons and their clusters causes the positive MR, and strongly affects the EL intensity and the emitted photon energy in the LEDEs.

2. Experimental

The bicrystals containing a single GB were cut from the $Zn_{1-x}Mn_xTe$ ($0 \leq x \leq 0.09$) ingots and from the ZnTe ingot for the reference. The EL was excited by a dc current with a density (J) between 0.08 and 1.7 A/cm^2 to prevent the heating of the samples. The EL emission was collected in the Faraday configuration in magnetic field up to 14 T directed parallel to the GB plane.

3. Results and discussion

The capacitance–voltage (C–V) and current–voltage (I–V) measurements for the ZnTe and $Zn_{1-x}Mn_xTe$ bicrystals revealed two symmetric electrostatic barriers with a height of $\approx 0.50 \pm 0.05$ eV joined back-to-back at the GB. Under a bias voltage of 4–6 V across the GB in both polarities, the I–V characteristics exhibit an electric breakdown. Simultaneously, the ZnTe and $Zn_{1-x}Mn_xTe$ LEDEs emit the intense EL light due to the lattice impact ionization in ZnTe and $Zn_{0.97}Mn_{0.03}Te$ and the direct impact excitation of Mn^{2+} ions by hot electrons (see lower part in Fig. 1).

Figure 1a presents EL spectrum of the ZnTe LEDE at 77 K which exhibits an EL peak at 2.325 ± 0.001 eV. Since this emission energy is close to the donor–acceptor pairs (DAP) transition energy in ZnTe:P (2.331 eV), we attribute the observed EL to the DAP recombination. The EL spectrum of the (Zn,Mn)Te LEDE at 77 K presented in Fig. 1b exhibits two EL bands, one at 2.338 eV resulting from the DAP recombination, and the other at 1.959 eV originating from the intra-Mn^{2+} transitions.

It was observed that the DAP EL emissions from the ZnTe and (Zn,Mn)Te LEDEs behave oppositely with increasing magnetic field. Figure 2a presents the magnetic field dependence of the integrated EL intensities normalized to the zero-field intensity at 4.2 K, $I_{\mathrm{n}}(B,T) = I(B,T)/I(0, 4.2\text{ K})$, of the (Zn,Mn)Te LEDE at 4.2 and 1.8 K, and that of the ZnTe LEDE at 4.2 K. It was found that at 4.2 K the $I_{\mathrm{n}}(B)$ of the ZnTe LEDE decreases from 1 to 0.28 when the applied magnetic field increases from 0 to 14 T, i.e. the measured EL intensity decreased by ≈ 3.6 times. At the same time the $I_{\mathrm{n}}(B, 4.2\text{ K})$ of the (Zn,Mn)Te increases from 1 to 17 in the same magnetic field range.

At 1.8 K the zero-field EL intensity $I_{\mathrm{n}}(0, 1.8\text{ K})$ is only 0.24. This means that in the absence of the magnetic field the EL intensity from the (Zn,Mn)Te LEDE *de-*

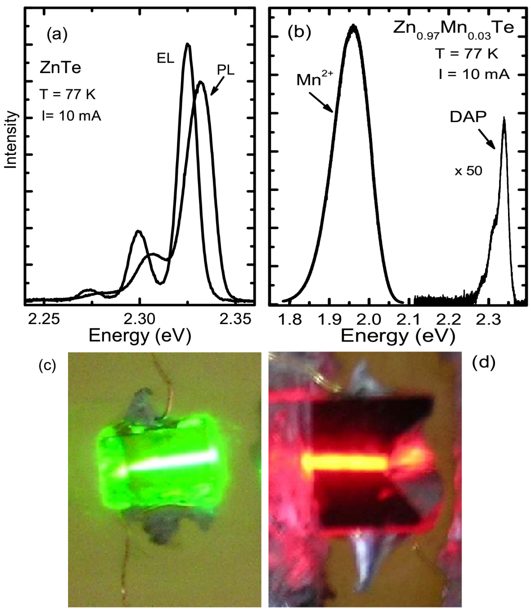

Fig. 1. (a) PL spectrum of ZnTe:P and EL spectrum of the ZnTe LEDE. (b) EL spectrum of the $Zn_{0.968}Mn_{0.032}Te$ LEDE at 77 K. Lower part shows photographs of the ZnTe (c) and $Zn_{0.968}Mn_{0.032}Te$ (d) LEDEs.

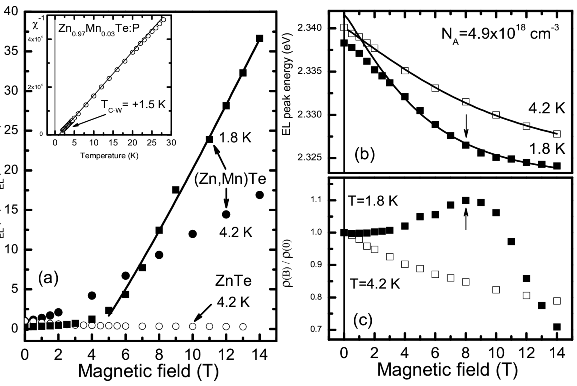

Fig. 2. (a) The normalized EL intensity as a function of magnetic field for the (Zn,Mn)Te LEDE at 4.2 K (full circles) and 1.8 K (full squares) and ZnTe LEDE at 4.2 K (open circles). Inset shows $1/\chi$ vs. T for $Zn_{0.97}Mn_{0.03}Te$:P. (b) EL peak energy vs. magnetic field. (c) The MR as a function of magnetic field at 1.8 K and 4.2 K.

creases by factor of ≈ 4.1 when the sample temperature is lowered from 4.2 K to 1.8 K. Application of an increasing external magnetic field increases the $I_{\mathrm{n}}(B, 1.8\ \mathrm{K})$ from 0.24 to 0.74 at $B_{\mathrm{c}} \approx 4$ T. Above 4 T, the $I_{\mathrm{n}}(B,\ 1.8\ \mathrm{K})$ sharply increases to 36.6 at $B = 14$ T, i.e. the measured EL intensity increases 152 times in the field range 0–14 T. These findings show that the normalized EL intensity exhibits a critical behavior in the magnetic field and it is described by a relation: $I_{\mathrm{n}}(B, 1.8\ \mathrm{K}) \sim |B - B_{\mathrm{c}}|^{\gamma}$ with $\gamma = 1.1 \pm 0.1$ for $B \geq 4$ T.

Figure 2b presents the magnetic field dependence of the EL peak energies of the (Zn,Mn)Te LED measured at 1.8 and 4.2 K, and the calculated energies based on the model presented in Ref. [1], $E(B) = E(0) + 0.8N_0\beta x S_0 B_{5/2}[5\mu_{\mathrm{B}}B/k_{\mathrm{B}}(T + T_0)]/2$, where $N_0\beta = -1.09$ eV is the exchange integral for the valence band [2], $x = 0.03$ is the molar fraction of Mn^{2+} ions, $E(0) = 2.3418\ (2.3401)$ eV, $S_0 = 0.55\ (0.46)$ is the effective Mn^{2+} spin, $T_0 = 9$ K (15 K) is an adjusting parameter at $T = 1.8$ K (4.2 K), $B_{5/2}$ is the Brillouin function for spin $S = 5/2$, the factor 0.8 describes the orbital quenching of the acceptor splitting [3]. Here, the donor splitting is neglected. It is seen that the measured peak energies at 4.2 K are in good agreement with the calculated one in the whole field range, whereas the experimental data at 1.8 K exhibit a marked departure from the theoretical curve in a field range $0 \leq B \leq 3$ T. Namely, the zero--field photon energy measured at 1.8 K is smaller than the calculated energy by 4.4 ± 1.5 meV, and the difference between the measured and calculated energies decreases to zero when the field exceeds 8 T. Moreover, the measured zero-field photon energy at 1.8 K is also smaller than that obtained at 4.2 K by ≈ 2.1 meV. These findings show that the binding energy of the phosphorus acceptors at 1.8 K increases in respect of that at 4.2 K, i.e. there is a formation of acceptor bound magnetic polarons (ABMPs) in (Zn,Mn)Te.

Further, the magnetic susceptibility (χ) measurements revealed that the Curie–Weiss temperature is positive, ferromagnetic, and equals $T_{\mathrm{CW}} = +1.5$ K (see inset in Fig. 2a). This indicates that at the phosphorus doping level of $N_{\mathrm{A}} = 4.9 \times 10^{18}\ \mathrm{cm}^{-3}$ the χ in (Zn,Mn)Te:P is enhanced, and the carrier-mediated Mn–Mn ferromagnetic (FM) interaction overcompensates the intrinsic antiferromagnetic superexchange. Thus we conclude that there is a coalescence of the isolated ABMPs resulting in ferromagnetic (FM) clusters in the (Zn,Mn)Te at 1.8 K.

Figure 2c shows the variation of the magnetoresistance, $\mathrm{MR}(B) = \rho(B)/\rho(0)$, with magnetic field at 1.8 and 4.2 K. It is seen that at 4.2 K the MR monotonically decreases with increasing field. It was found that the MR(4.2 K) is proportional to the Zeeman shift of the EL emission energy, $\Delta E_{\mathrm{Z}} = E(0) - E(B)$, by a relation $\mathrm{MR}(B) = 0.98 - K \times \Delta E_{\mathrm{Z}}(B)$ where $K = 10.24\ \mathrm{eV}^{-1}$ for the fields $B \geq 1$ T.

However, at 1.8 K the MR does not correlate with the ΔE_{Z}. First it weakly decreases passing a minimum MR = 0.998 at $B \approx 1.5$ T then it *increases* with field reaching a maximum MR = 1.10 at $B \approx 8$ T. Above 8 T the MR abruptly decreases. These findings indicate that the 1.8 K MR contains both negative and positive components. The negative component arises from the exchange induced decrease of the acceptor binding energy, and from the suppression of the fluctuation of magnetization. The positive component originates from the suppression of the hopping conduction of the carriers between the isolated BMPs and/or FM clusters. Since the isolated BPMs are stable only at low fields $B \leq 3$ T, the results presented in Fig. 3c at 1.8 K show that the FM clusters are stable up to a field of 8 T, that causes the domination of the positive MR in this field range. Above 8 T, the ΔE_Z which is proportional to the sample magnetization saturates (Fig. 3b). This leads to disappearance of the FM clusters together with their conduction. As a result, the negative MR component dominates at higher fields.

4. Conclusion

We have fabricated the LEDEs using the ZnTe and (Zn,Mn)Te bicrystals. The (Zn,Mn)Te LEDEs produce red and green emission originating from the intra *d*-shell transitions in the Mn^{2+} ions and the DAP recombination, respectively. The presence of the ABMPs and their clusters near the Curie–Weiss temperature in (Zn,Mn)Te was observed. It induced a critical behavior of the magnetic field dependence of the green EL intensity and positive MR in the LEDEs.

Acknowledgments

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