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Magnetism and carrier modulation in (Ga,Mn)As/organic-dye hybrid devices

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We present the manipulation of magnetic and electrical properties of (Ga,Mn)As via the adsorption of dye-molecules as a step toward the realization of light-controlled magnetic-semiconductor/dye hybrid devices. A significant lowering of the Curie temperature with a corresponding increase in electrical resistance and a higher coercive field is found for the (Ga,Mn)As/fluorescein system with respect to (Ga,Mn)As. Upon exposure to visible light a shift in Curie temperature toward higher values as well as a reduction of the electrical resistance and the coercive field can be achieved. This points toward a hole quenching effect at the molecule-(Ga,Mn)As interface which is susceptible to light exposure. © *2011 American Institute of Physics*. [doi:10.1063/1.3543843]

One of the most striking characteristic of the ferromagnetic semiconductor (Ga,Mn)As (Ref. 1) is the possibility to tune its magnetic properties via the manipulation of the carrier density.[2] Using metallic gates in a classic field effect transistor geometry allows for the continuous tuning of the magnetization in the absence of magnetic fields.[3] Moreover, ferroelectric gates can modulate the carrier density in a nonvolatile way.[4]

In this study we present the hole quenching effect of the adsorption of light-sensitive molecular layers on the magnetic and electrical properties of (Ga,Mn)As/GaAs Hall bar devices. We also show, as a proof of principle, the possibility to influence this hole quenching process by light excitation as an important step toward magnetic applications exploiting molecule-dependent, photon-controlled carrier modulation.

The two (Ga,Mn)As/GaAs materials employed are designated A and B, for details regarding the epitaxial growth, see Refs. 5 and 6. Sample A has a Curie temperature T_c of 68 K, a thickness of 50 nm, and a Mn concentration of 8%. For sample B the values for the same parameters are 48 K, 40 nm, and 5%, respectively. The molecular layers were adsorbed by immersing the (Ga,Mn)As films in an aqueous 2 mM solution of the molecules. Prior to the immersion in the solution for about 12 h the films are treated with an HF containing etch mixture (original etch mixture: AF 87.5–12.5 VLSI Selectipur, diluted 1:100 in water) for approximately 10 s to clean the surface. Magnetotransport measurements performed before and after the HF treatment confirm that the etching procedure does not modify the properties of the films. The light source employed in the illumination experiments is a standard HBO Hg lamp. No light-induced effects have been observed in the magnetotransport properties in the as-grown films.

Fluorescein (see Fig. 2) is thought to interact with the (Ga,Mn)As surface mainly via the carboxylic group as proposed in the literature for the chemisorption of carboxyl-terminated molecules on GaAs/(Ga,Mn)As multilayers[7] and for the adsorption of fluorescein and other aromatic acids on semi-insulating and metallic surfaces.[8] The x-ray photoelectron spectra (XPS) of sample A before and after fluorescein adsorption at room temperature are shown in Figs. 1(a) and 1(b), respectively. The major changes in the spectra related to the adsorption of fluorescein occur at lower binding energies with respect to the Ga 3*d* peak at 19.6 eV [the shoulder toward higher binding energies (21.3 eV) accounts for the presence of gallium oxide].[9] These energies correspond to the valence band states in bulk GaAs(001) (Ref. 10) and by analogy also to the valence band of its *p*-doped variant (Ga,Mn)As. The distinct increase of the spectral intensity in this region indicates the appearance of occupied electronic levels in the energy range of the (Ga,Mn)As valence band states upon adsorption of fluorescein. This lineshape most likely reflects changes occurring in the (Ga,Mn)As states due to the interaction with the adsorbates together with the occupied states of the molecule. The appearance of populated levels at these relatively low binding energies could point at an effective hole quenching in the (Ga,Mn)As substrate that according to the theory of hole mediated ferromagnetism[2] would lead to the weakening of the ferromagnetic interaction.

Presented in Fig. 2 is the temperature dependence of the electrical resistance in the absence of light showing the characteristic peak (inset) indicative of the Curie temperature[11]

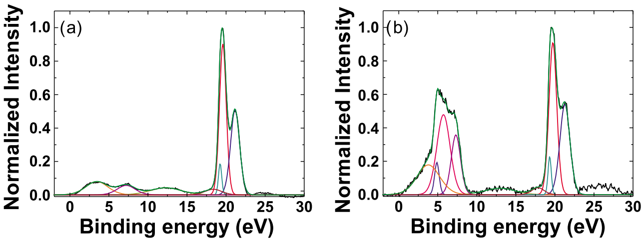

FIG. 1. (Color online) Normalized XPS spectra of (Ga,Mn)As at low binding energies before (a) and after (b) adsorption of fluorescein.

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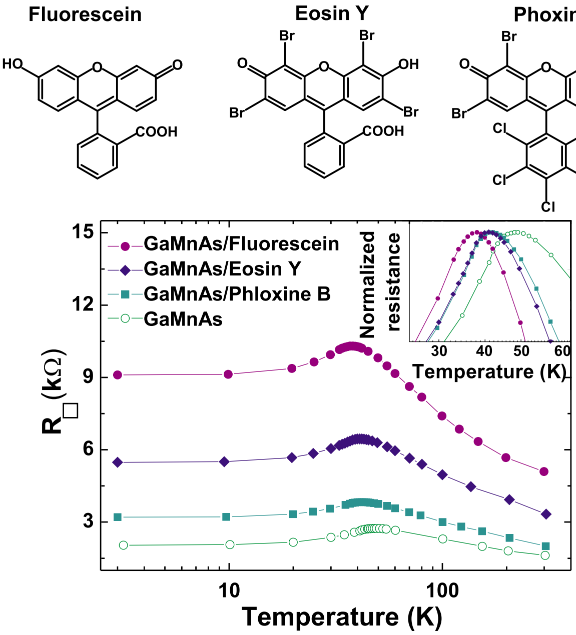

FIG. 2. (Color online) Effect of fluorescein and its derivatives eosin Y and phloxine B on the electrical properties of sample B in the absence of light. Inset: normalized resistance peaks.

for the interaction between (Ga,Mn)As (sample B) and different fluorescein derivatives. In line with the picture of a hole compensation mechanism derived from the XPS spectra an increase in the electrical resistance and a shift of the resistance peak toward lower temperatures is observed for all the molecules, fluorescein producing the largest effect. The two fluorescein derivatives investigated are eosin Y (diamonds) and phloxine B (squares) that differ from fluorescein by additional electronegative bromide and chlorine groups (see Fig. 2). The presence of electronegative groups is expected to hinder the hole quenching capability of the molecule and is in agreement with our experiments. The adsorption of eosin Y produces a smaller decrease in the Curie temperature and a reduced increase in the electrical resistance with respect to fluorescein. An even less efficient hole quenching is observed for phloxine B where a slightly higher Curie temperature and a smaller resistance increase is observed with respect to eosin Y.

In Fig. 3 the coercive fields H_{c1} (circles) and H_{c2} (squares) at a temperature of 2 K for (Ga,Mn)As and (Ga,Mn)As/fluorescein as a function of the direction of the applied magnetic field with respect to the [110] uniaxial easy axis direction are displayed. The magnetic anisotropy landscape given by the coercive fields map shows a rectangular shape with its long axis parallel to the [110] direction for H_{c1} and H_{c2} approaching the [110] and $[1\bar{1}0]$ axes which is typical for as-grown (Ga,Mn)As materials.[6,14] The values of the coercive fields were obtained by planar Hall effect measurements.[14] After adsorption of the molecules an overall increase of the coercive fields in all directions can be observed in agreement with the lower value of T_c as it has been reported by other authors.[12] A change in the magnetic anisotropy upon fluorescein adsorption is evidenced in a modest strengthening of the uniaxial easy axis ([110] axis, 0° direction) where the increase in coercivity is larger than along the uniaxial hard axis direction ($[1\bar{1}0]$ axis, 90° direction). The ratio $H_c[1\bar{1}0]/H_c[110]$ decreases from 0.75 for the as-grown (Ga,Mn)As to 0.71 for (Ga,Mn)As/fluorescein, a 5% shift in favor of the [110] easy axis direction. It has been shown both theoretically and experimentally that not only the Curie temperature but also the magnetic anisotropy in (Ga,Mn)As is susceptible to variations of the carrier density.[2,3,13] Experimental work shows that the uniaxial term is the most sensitive to variations in the carrier concentration,[3] thus, our observation of a change in the uniaxial anisotropy component with respect to the biaxial component upon changes in the carrier concentration is clearly in line with results in the literature. The general increase of coercivities in all directions can be discussed in terms of micromagnetic models. Since the magnetization reversal in this material is known to occur by domain wall motion[6] a common expression for the domain wall pinning energy density (ϵ) can be included in this analysis:[14] $\epsilon = (\mathbf{M}_2 - \mathbf{M}_1) \cdot \mathbf{H}_c$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the initial and final magnetization vectors and $\mathbf{H}_c$ is the coercive field. Keeping in mind the small changes in the magnetic anisotropy after the adsorption of fluorescein we can assume that ϵ remains approximately constant for every direction. Therefore, according to the above expression a general increase in $\mathbf{H}_c$ could be directly related to a decrease in the absolute magnetization value M that is in agreement with the notion of a weakened ferromagnetic interaction given by the smaller value of Curie temperature after adsorption of fluorescein.

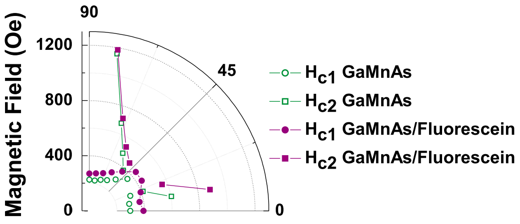

FIG. 3. (Color online) Coercive fields H_{c1} (circles) and H_{c2} (squares) as a function of the angle of the applied magnetic field with respect to the [110] direction at 2 K for sample B.

The shift in T_c upon adsorption of fluorescein for sample A is 18 K as shown in Fig. 4(a) and like in the case of sample B it is accompanied by the corresponding increase in resistance (inset). It is useful to observe that in the present case these two quantities can be linked together by the well known expression $T_c \propto p^{1/3}$ provided by mean field theory,[2] where p is the hole carrier density. The ratio $T_c^{\text{(Ga,Mn)As}}/T_c^{\text{(Ga,Mn)As/Fluorescein}} \sim 1.36$ is in good agreement with the ratio of the cubic root of the inverse resistances at 3 K (assumed to be proportional to p) which has a value of approximately 1.35. A similar relation is found for sample B.

In the following we show results which prove that the (Ga,Mn)As-fluorescein interaction introduces an additional degree of freedom for magnetic manipulation that can be controlled by light. In our approach we do not intend to influence the magnetic properties by direct interaction between (Ga,Mn)As and light[15] but instead modulate the hole quenching capability of the fluorescein molecules as a sort of light-regulated gate.

A distinct shift of the resistance peak toward higher temperatures during illumination can be observed in Fig. 4 (top) which is accompanied by a decrease in the value of the resistance (inset). In addition, a decrease in the value of the

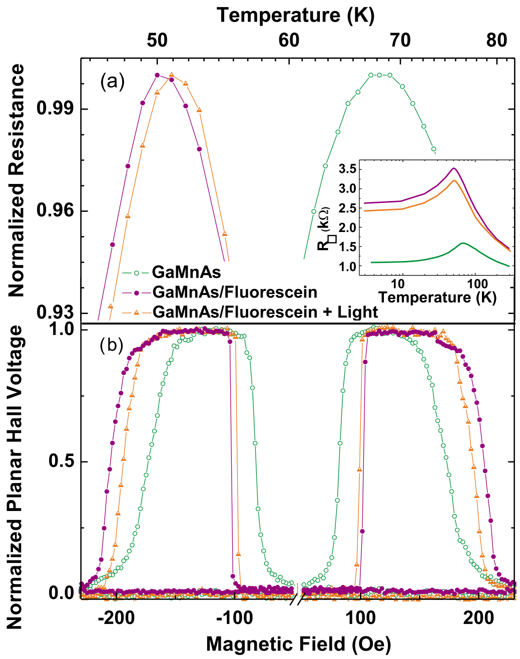

FIG. 4. (Color online) Effect of light exposure on functionalized (Ga,Mn)As devices. (a) Normalized resistance peaks in the temperature dependencies of the electrical resistance (inset). (b) Magnetic field dependence of the normalized planar Hall voltage.

coercive field is observed in the field dependence of the planar Hall voltage as shown in Fig. 4 (bottom). Thus, the illumination effect is inverse to the molecule adsorption effect and according to our discussion we attribute the effect of light to an increase of hole carriers in (Ga,Mn)As. The molecule ground state is expected to be located close to the (Ga,Mn)As valence band in order to be able to effectively interact with the holes in these levels. Upon light absorption it is possible that a fraction of the electrons that were interacting with the valence band are now being promoted to the excited state of the molecule and are no longer able to contribute to the hole compensation producing a small increase in the hole concentration and an increase in the Curie temperature.

In conclusion, the results presented here demonstrate the varying effectiveness of different molecular layers in quenching hole carriers in (Ga,Mn)As and in turn changing its magnetic properties. Most importantly this study demonstrates that this interaction can be modified by optical excitation of the molecular layers as a step toward light-controlled ferromagnetism in (Ga,Mn)As/dye devices.

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