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Second harmonic generation probing of MnAs/Si(111) heterostructures

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Abstract. Ferromagnetic-semiconductor heterostructures MnAs/Si(111) have been studied by using the optical second harmonic generation and the linear magneto-optical Kerr effect. Magnetic and crystallographic contributions to the SHG are separated on the base of rotational anisotropy method. Magnetic hysteresis loops measured by the SHG and the Kerr effect exhibit different behavior due to a different sensitivity of two optical techniques to the interface, surface and bulk properties.

Introduction

Magneto-electronics based on new types of nanostructures is rapidly developing brunch of technique. Ferromagnetic-semiconductor heterostructures are a subject of intensive investigations due to their great potential for new optical and electronic devices, such as nonvolatile magnetic memory monolithically integrated with Si or GaAs in one chip, magneto-optical memory, magnetic sensors coupled with semiconductor circuits and spin-polarized transport devices [1]. For this kind of devices it is important to monitor the interface and bulk magnetic properties of the ferromagnetic films.

For centrosymmetric media the second harmonic generation (SHG) is only allowed in the electric-dipole approximation for surfaces and interfaces, where the space-inversion-symmetry operation is broken [2]. Thus SHG is expected to be sensitive to a few monolayers at surfaces and interfaces. It has been experimentally shown that the source of magnetization-induced SHG signal for Co films is bound to the interface region of less than 6 monolayers [3]. On the other hand, the linear magneto-optical Kerr effect (MOKE) is formed in the thickness range of the penetration depth of light $\alpha = \lambda/(4\pi k)$ [4], where λ is the wavelength of light and k is the absorption coefficient. We suggest that SHG and MOKE methods can provide complementary results due to their different probing regions.

The nonlinear optical polarization excited in a noncentrosymmetric magnetic medium can be written in the electric-dipole approximation as [5]

$$P_i^{2\omega} = \varepsilon_0 \chi_{ijk}^{(2)} E_j E_k + \varepsilon_0 \chi_{ijkl}^{(3)} E_j E_k M_l,$$

where E_j and E_k are the incident optical electric fields at the fundamental frequency ω and $\mathbf{M}$ is the spontaneous magnetization. The nonlinear tensors $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$ describe the crystallographic and magnetic parts of $\mathbf{P}^{2\omega}$, respectively. These tensors are complex in the absorption region of crystals, what allows interference between crystallographic and magnetic contributions to SHG.

In the paper we present results on the SHG and MOKE studies MnAs/Si(111) heterostructures.

1 Experiment and samples

Below the critical temperature $T_C = 45^\circ \text{C}$, MnAs has the hexagonal structure of NiAs-type (point group $6/mmm$) and ferromagnetic state with an “easy plane” type of magnetic anisotropy coinciding with the (0001) crystal plane. At T_C , a first order phase transition occurs to the paramagnetic structure of MnP-type having the point group mmm . Heterostructures $\text{CaF}_2/\text{MnAs}/\text{Si}(111)$ have been grown by the molecular-beam-epitaxy method. After standard chemical cleaning, silicon substrates were loaded into the growth chamber and cleaned thermally at 1250°C in ultra high vacuum. This procedure provides atomically cleaned $\text{Si}(111)$ surfaces with a 7×7 superstructure. The thickness of MnAs films was in the range of 12–400 nm. A cap layer of CaF_2 (thickness 4–12 nm) was used for protection. The crystalline quality of Si substrates and the growth of ferromagnetic films and cap layers was monitored *in situ* by reflection high energy electron diffraction. For the SHG and MOKE experiments, we used a Ti:sapphire laser, with 100 fs pulse width at 80 MHz repetition rate (see Ref. [5]). All experiments have been done in reflection at a wavelength of 800 nm and at an angle of incidence of 45° .

2 Results and discussion

In Fig. 1 the anisotropy of SHG in $\text{CaF}_2(4 \text{ nm})/\text{MnAs}(35 \text{ nm})/\text{Si}(111)$ structure is shown for the two directions of $\mathbf{M}$ in the plane of incidence (longitudinal geometry) as a function of crystal rotation angle φ around the sample surface normal. Measurements have been done for the *P*-input polarization of the fundamental light and *S*- (Fig. 1(a)) and *P*- (Fig. 1(b)) output polarizations of the SH light. In case of the *PS* combination of polarizations the magnetic contribution to SHG survives and it is possible to separate it from the crystallographic one. The experimental data are well described by the equation $I^{2\omega} = [A \cos(3\varphi) + B]^2$, where A and B are the constants describing the anisotropic and isotropic contributions to the SHG, respectively. In case of *PS* combination of polarizations B depends linearly on M , but if the combination is *PP* then B is independent on M and is purely crystallographic. The best fit is shown by the solid line for $+M$ and by the dashed line for $-M$ in Fig. 1(a). The anisotropic contribution to the SHG arises partially from the MnAs/Si interface and the Si(111) surface characterized by the point group $3m$ (about 30% of Si surface was not covered by MnAs during deposition). For this polar group there are four independent components of $\chi_{ijk}^{(2)}$ ($xxx = -xyy = -yyx = -yxy$, $yyz = xxz = xzx = yzy$, $zxx = zyy$, zzz [6]). The isotropic magnetic contribution to SHG is most probably related to the CaF_2/MnAs magnetic interface. This interface dominates over MnAs/Si interface due to a strong absorption of the pump beam in the MnAs film ($k = 2.8$ at $\lambda = 800 \text{ nm}$ [7]).

Fig. 2 shows the rotational anisotropy of the SH *P*-polarized intensity for the $\text{CaF}_2(8 \text{ nm})/\text{MnAs}(70 \text{ nm})/\text{Si}(111)$ structure for the *P*-polarized fundamental light. In this structure MnAs film is sufficiently thick to cancel the contribution from MnAs/Si interface. Thus in this case the SHG may arise only from CaF_2/MnAs interface. Possible polar group for this interface is $6mm$, therefore the SHG should exhibit an isotropic behavior. In the transversal geometry ($\mathbf{M}$ is perpendicular to the plane of incidence) there is a noticeable magnetic contribution to the SHG (see Fig. 1(a)). In the longitudinal geometry magnetic contribution to SHG vanishes, in accordance with the symmetry properties of $\chi_{ijkl}^{(3)}$.

For the azimuthal angle position of crystal $\varphi = 210^\circ$, where the magnetic contrast has a maximum value (see Fig. 1(a)), we measured the SHG and MOKE hysteresis loops shown in Fig. 3(a) for $\text{CaF}_2(4 \text{ nm})/\text{MnAs}(35 \text{ nm})/\text{Si}(111)$ structure. There is a pronounced difference between the coercive fields measured by the two methods. This difference is

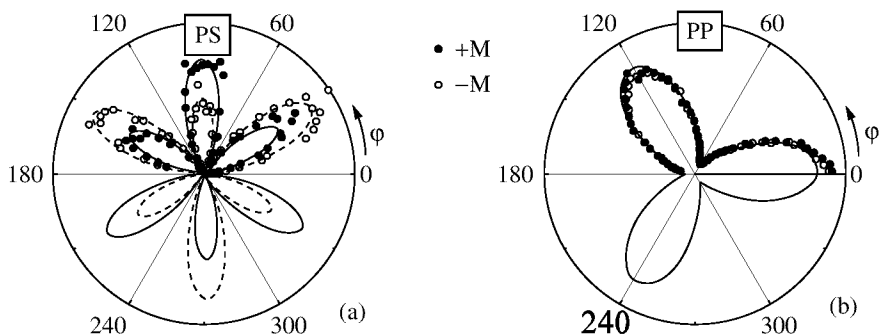


Fig. 1. Rotational anisotropy of the SH intensity in longitudinal geometry for $\text{CaF}_2(4\text{ nm})/\text{MnAs}(35\text{ nm})/\text{Si}(111)$ structure for *PS* (a) and *PP* (b) input-output combinations of polarizations.

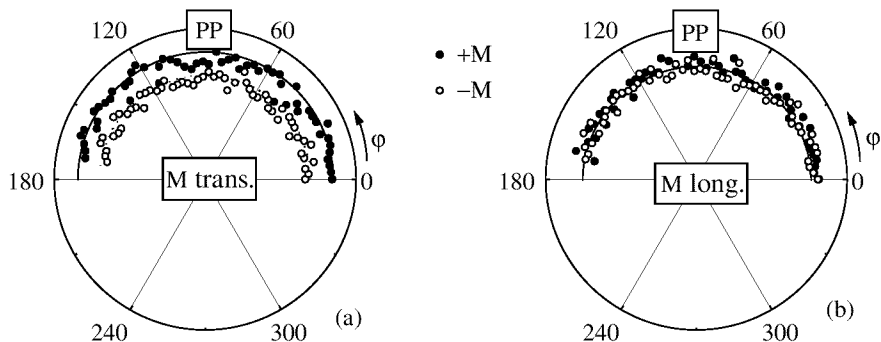


Fig. 2. Rotational anisotropy of SH intensity in (a) transversal and (b) longitudinal geometries for $\text{CaF}_2(8\text{ nm})/\text{MnAs}(70\text{ nm})/\text{Si}(111)$ structure for *P*-input and *P*-output polarizations.

stronger for $\text{CaF}_2(8\text{ nm})/\text{MnAs}(70\text{ nm})/\text{Si}(111)$ structure (Fig. 3(b)). Moreover, the SHG hysteresis loop is shifted in the direction of positive value of magnetic field.

In conclusion, $\text{CaF}_2/\text{MnAs}/\text{Si}(111)$ heterostructures have been studied by using the two complementary magneto-optical techniques. Anisotropic pattern of SH rotational

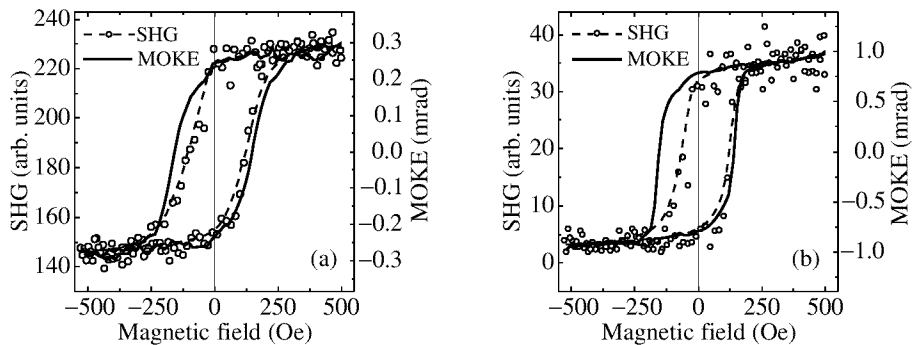


Fig. 3. Comparison of magnetic hysteresis loops measured using the SHG (dots and dashed lines are guide for eye only) and the Kerr effect (solid lines) for (a) $\text{CaF}_2(4\text{ nm})/\text{MnAs}(35\text{ nm})/\text{Si}(111)$ and (b) $\text{CaF}_2(8\text{ nm})/\text{MnAs}(70\text{ nm})/\text{Si}(111)$ structures.

anisotropy has been observed for $\text{CaF}_2(4 \text{ nm})/\text{MnAs}(35 \text{ nm})/\text{Si}(111)$ structure, whereas for $\text{CaF}_2(8 \text{ nm})/\text{MnAs}(70 \text{ nm})/\text{Si}(111)$ it was found to be isotropic. In both cases the crystallographic and magnetic contributions to the SHG have been separated on the base of model calculations. It was found that the SHG hysteresis loop differs from the MOKE one for the structures under study. Taking into account a sensitivity of the SHG and MOKE to the CaF_2/MnAs interface and the bulk of MnAs films, respectively, we can make conclusions about difference in magnetic properties of probed areas of the ferromagnetic films.

Acknowledgements

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