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ON THE POSSIBLE ORIGIN OF LARGE VALUE OF MAGNETIC ANISOTROPY CONSTANT IN PbSnMnTe MIXED CRYSTALS*

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In PbSnMnTe crystals with the concentration of conducting holes of the order of 10^{21} cm^{-3} , the magnetic anisotropy constant K measured in ferromagnetic resonance experiments is nearly one order of magnitude larger than expected. In the present paper we investigate a contribution ΔK to the anisotropy constant, origin of which is the s - d interaction and the spin-orbit coupling.

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The free energy of a magnetic ion in a cubic crystal, as the function of the magnetization M has the following form:

$$F\{M\} = F_0 + K(n_x^2 n_y^2 + n_y^2 n_z^2 + n_z^2 n_x^2). \quad (1)$$

In Eq. (1) the term F_0 does not depend on $n_\alpha = M_\alpha/M$ ($\alpha = x, y, z$), i. e. on the direction of the magnetization. The anisotropic part is described by the second term and the strength of the anisotropy is given by the anisotropy constant K .

Usually K is of the order of 10^{-7} - 10^{-6} eV [1]. However, in crystals $\text{Sn}_{1-x}\text{Mn}_x\text{Te}$ and $\text{Pb}_{0.28-x}\text{Sn}_{0.72}\text{Mn}_x\text{Te}$ ($0.01 < x < 0.04$) with very large concentration of conducting holes (of the order of 10^{21} cm^{-3}) the value of K measured in ferromagnetic resonance experiments is nearly one order of magnitude larger [2].

In this paper we investigate a contribution to the magnetic free energy, origin of which is the interaction between the magnetic ion and the conduction band holes (the s - d interaction). It turns out that if the concentration of the free carriers is large, the presence of the s - d interaction may result in the increase in the anisotropy constant K .

The plan of our approach to the problem is following. We consider a perfect crystal containing one magnetic ion placed at the origin of the coordinate system. We calculate the free energy of such a system as the function of the magnetic ion

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spin $\mathbf{S} = (S^x, S^y, S^z)$, which is treated as the classical Heisenberg vector. The term of fourth order in the Taylor expansion of the free energy in powers of S^α should give us the information about the contribution ΔK to the anisotropy constant caused by the s - d interaction.

The Hamiltonian of our system has the following form:

$$H = H_0 + H_{s-d} = H_0 + \mathbf{S} \int d^3r J(\mathbf{r}) s(\mathbf{r}). \quad (2)$$

H_0 is the Hamiltonian of the translationally invariant perfect crystal. The second term in Eq. (2) describes the s - d interaction with $s(\mathbf{r})$ being the spin density operator of the conducting holes and $J(\mathbf{r})$ the coupling between the magnetic ion and the valence band holes.

In the language of the second quantization the Hamiltonian reads

$$H = \sum_{nk} \varepsilon_{nk} a_{nk}^\dagger a_{nk} + \sum_{nk} \sum_{n'k'} \mathbf{S} \cdot \mathbf{Q}_{nk,n'k'} a_{nk}^\dagger a_{n'k'}. \quad (3)$$

The operator a_{nk}^\dagger creates a particle in an eigenstate $\begin{pmatrix} \phi_{+nk}(\mathbf{r}) \\ \phi_{-nk}(\mathbf{r}) \end{pmatrix}$ of H_0 with the energy ε_{nk} . The quantities $\mathbf{Q}_{nk,n'k'}$ are defined as follows:

$$\mathbf{Q}_{nk,n'k'} = \int d^3r J(\mathbf{r}) (\phi_{+nk}^*(\mathbf{r}), \phi_{-nk}^*(\mathbf{r})) \boldsymbol{\sigma} \begin{pmatrix} \phi_{+n'k'}(\mathbf{r}) \\ \phi_{-n'k'}(\mathbf{r}) \end{pmatrix}, \quad (4)$$

where $\boldsymbol{\sigma} = (\sigma^x, \sigma^y, \sigma^z)$ are the Pauli matrices. Because the Hamiltonian H is a bilinear form in a_{nk} and a_{nk}^\dagger , it is very easy to write the formal expression for the magnetic free energy $F\{\mathbf{S}\} = -\frac{1}{\beta} \ln(\text{Tr} e^{-\beta H}) = -\frac{1}{\beta} \ln \det A$ [3], where the matrix A is defined in the following way:

$$A_{\tau nk; \tau' n'k'}(\mathbf{S}) = \left(\frac{\partial}{\partial \tau} + \varepsilon_{nk} - \mu \right) \delta(\tau - \tau') \delta_{nn'} \delta_{kk'} + \mathbf{S} \cdot \mathbf{Q}_{nk,n'k'} \delta(\tau - \tau'). \quad (5)$$

τ is the continuous variable from the interval $(0, \beta \equiv 1/kT)$ and μ is the chemical potential.

The above formulation provides a convenient starting point for the Taylor expansion of $F\{\mathbf{S}\}$ in powers of \mathbf{S} . We are interested in the fourth-order term which may be written in the form

$$\delta^{(4)} F\{\mathbf{S}\} = \frac{1}{4} \sum_{\alpha\beta\gamma\lambda} S^\alpha S^\beta S^\gamma S^\lambda \times \sum_{m=-\infty}^{\infty} \sum_{n_i k_i} Q_{n_1 k_1, n_2 k_2}^\alpha \cdots Q_{n_4 k_4, n_1 k_1}^\lambda \prod_i \frac{1}{i\omega_m - (\varepsilon_i - \mu)}, \quad (6)$$

where $\varepsilon_i \equiv \varepsilon_{n_i k_i}$, $i = 1, \dots, 4$ and $\omega_m = (2m + 1)\pi/\beta$. Let us apply the general formula (6) to the case of PbSnTe. We know that for the samples with concentration of holes of the order of 10^{21} the majority of the free carriers is grouped in the second valence band Σ or more precisely in twelve equivalent valleys of this band. Thus we assume that we deal only with this band and the presence of other bands is neglected.

Secondly, we suspect that $\mathbf{Q}_{nk,n'k'}$ should be approximately constant for small values of $k - k'$. Consequently, we assume that $\mathbf{Q}_{nk,n'k'} = \mathbf{Q}_{nk\Sigma, n'k\Sigma} = \mathbf{Q}_{n,n'}$

if k and k' belong to the same Σ valley. We suspect also that $Q_{nk,n'k'}$ vanish if the distance between k and k' grows, therefore the matrix elements between different Σ valleys are neglected.

After assumption concerning the behaviour of $Q_{nk,n'k'}$, in order to perform the summation over k_i in Eq. (6) we must introduce some energy cut-off parameter E , because the summation over k_i is limited to one Σ valley. In the following we treat E as the fitting parameter. It cannot be much larger than the chemical potential $\mu \approx 200$ meV, because the energy distance to other Σ bands is of the order of 500 meV.

We calculate the quantities Q^α using the wave functions derived by Pankratov and Volkov [4] in their so-called p -model. Finally, we obtain

$$\Delta K = 16 \left(\frac{n\Omega S\alpha \sin \gamma}{16\mu} \right)^4 \mu F \left(\frac{\mu}{E} \right), \quad (7)$$

where

$$F(z) = z^{-3} \int_0^z dx \sqrt{x} \left(2 + \sqrt{x} \ln \frac{1-\sqrt{x}}{1+\sqrt{x}} \right) \left[\left(2 + \sqrt{x} \ln \frac{1-\sqrt{x}}{1+\sqrt{x}} \right)^2 - \pi^2 x \right], \quad (8)$$

Ω is the volume of the elementary cell, $\alpha = (\Omega/V)\bar{\alpha}$ and $\bar{\alpha}$ is one of the exchange integrals between magnetic ion and the valence band holes of PbTe (that one described by the functions of s -type) and is of the order of 300 meV [5]. $\sin \gamma$ describes the spin-orbit coupling for the valence band of PbTe and it is approximately equal to 0.64 [4]. Taking the concentration of holes $n = 10^{21} \text{ cm}^{-3}$, $S = \frac{5}{2}$ and $E = 350$ meV, we obtain $\Delta K = -6 \times 10^{-3}$ meV which corresponds quite well to the experimental value $K = -2.5 \times 10^{-3}$ meV for PbSnMnTe [2].

Although the agreement between theory and experiment seems to be very good, one should not take Eq. (7) too quantitatively due to the assumed approximations. However, even if other factors in Eq. (7) might were over- or underestimated, the n^4 dependence of ΔK should retain (we neglect here the changes of μ with n), which may be verified experimentally.

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