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Non-equilibrium phase transition into ferromagnetic semiconductor nanofilms in an electric field

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ABSTRACT

The two-band Hubbard model is studied using electric-field heating of the electronic and magnetic subsystems of magnetic semiconductors. The sample $EuO_{1-\delta}$ is described using the experimentally observed significant dependence of the magnetization on the electric field and the S- or N-shaped current-voltage characteristics. It is shown that for thin films, smallest thickness may impair the stationary mode and the emergence oscillations of the current or the voltage. The length-to-thickness ratio of the film determines the period of this oscillation.

1. Introduction

Through a semiconductor whose electrical resistivity rapidly decreases with increasing temperature, the electric current is accompanied by a self-heating process, where the internal temperature of the sample is different from the temperature of its surface. In conditions such as homogeneous heating of the sample (over the thickness), the internal temperature is determined from the current strength and the applied voltage in accordance with the condition of thermal equilibrium:

$$C\frac{dT}{dt} = JU - \frac{\lambda(T-T_0)}{h}S \quad (1)$$

where t is time, U is the applied voltage, J is the current strength, T is the temperature inside the sample, T0 is the external temperature, C and λ are the heat capacity and the thermal conductivity of the sample, respectively, and S is the total square of the side surfaces of the sample. As shown in [1–3], the stationary current-voltage characteristics (CVC) of semiconductors of various forms can be S- or N-shaped. This shape indicates the formation of the bistable state (i.e., “cold” and “hot” phases).

A special and theoretically no studied by situation arises in magnetic semiconductors when the ferromagnetic metal-paramagnetic semiconductor magnetically transforms (see, e.g., [3–5]). For these substances, a significant and sharp change in the electrical conductivity ($\sigma(T)$) is observed near the magnetic phase transition temperature (thin film $EuO_{1-\delta}$, lanthanum manganite, etc.), which indicates a strong relation between the electron and the magnetic subsystems. Previously, it was shown that magnetic semiconductors that are heated by the electric field form magnetic subsystems because of the electron–magnon interaction (i.e., the concept of "hot magnon" [6]). In this work, the dependence of the magnetization on the electric field was obtained and was qualitatively consistent with the experiment. However, this approach is limited to low temperatures and inapplicable around the Curie temperature, where there is actually a sharp increase in electrical conductivity. Moreover, the experimental dependence σ(T0, U) and the current–voltage characteristics indicate that the heating by an external electrical field affects the magnetic subsystem and the electron subsystem [9].

2. Description of the model and the calculation

The effects of electric-field heating on the electron and magnetic subsystems of ferromagnetic semiconductors are examined using thin films of $EuO_{1-\delta}$. The electronic spectrum of this compound can be modeled based on the ab initio calculation of the density of EuO states (i.e., without oxygen vacancies) [7]. According to [7], the chemical potential of EuO is located between the valence and conduction bands, which are formed mainly d states. These two bands are separated by a wide energy gap whose width is - 3 eV. Filled energy levels of a 4f7-multiplet are located in the energy gap. The chemical potential of EuO is located between

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the 4f7-multiplet and the bottom of the conduction band. The energy levels of a multiplet merge into an energy region of width 0.6 eV [7].

To study the electronic and magnetic subsystems of $EuO_{1-\delta}$ and the analogous ferromagnetic semiconductors, we use the Hubbard model for two bands

$$H = H_{ff} + H_{dd} \tag{2}$$

where the Hamiltonians H_{ff}, H_{dd}, which have identical structures, are the Hamiltonian of non-interacting electrons in an external electric field and the Hamiltonian of the Hubbard repulsion at the site, respectively, which are recorded by a Fourier transform of the operators of the charge and the spin density (see, e.g., [8,10]), respectively. As in [8,10], we use the Laplace transforms and write the statistic sum of the system of electrons in the following form

$$Z = Sp\iint \exp\{-\beta U^{-1}\sum_{\nu}(|\xi_{\nu}|^2+|\eta_{\nu}|^2)\}T_{\tau}\{\exp[-\beta H(V)]\}(d\xi d\eta). \tag{3}$$

Here, the effective Hamiltonian $H(V)$ has the form

$$H(V) = \sum_{k,l,\sigma}\varepsilon_{k,l}a^{+}_{k,l,\sigma}a_{k,l,\sigma} + \sum_{k,q,l,\sigma}V_{q,l,\sigma}a^{+}_{k,l,\sigma}a_{k+q,l,\sigma} + \sum_{k,q,l}(V^{(+)}_{q,l}a^{+}_{k,l,\downarrow}a_{k+q,l,\uparrow} + \text{h.c.}),$$

T_τ is the ordering operator on the Matsubara time τ; $V^{(+)}_{l,\nu} = (\xi_{l,\nu,x}+i\xi_{l,\nu,y})/\sqrt{2}$; $V_{q,l,\sigma} = \xi_{l,\nu,z}+i\sigma\eta_{l,\nu}/2$; $\nu = (\boldsymbol{\nu},\tau)$; $c^{(l)} = [U^{(l)}T]^{1/2}$; $\eta_{l,\nu} = c^{(l)}\sum_q e^{iq\nu}\eta_{l,q}$; $(d\boldsymbol{\xi}\, d\eta) = \prod_{l,\gamma}\left(d\xi_{l,0,\gamma}d\eta_{l,0}/\pi\prod_{q\neq 0, j=1,2}d\xi^{(j)}_{l,q,\gamma}d\eta^{(j)}_{l,q}/\pi\right)$; $\sum_\nu(\ldots) = T\int_0^\beta d\tau\sum_{\boldsymbol{\nu}}(\ldots)$; $\boldsymbol{\xi}_{l,\nu} = c^{(l)}\sum_q \exp(iq\nu)\boldsymbol{\xi}_{l,q}$; $l = f,d$; $q = (\mathbf{q},\omega_{2n})$; $k = (\mathbf{k},\omega_{2n+1})$; ω_{2n} and ω_{2n+1} are the Bose and Fermi Matsubara frequencies; $U^{(l)}$ is Hubbard's constant; $a^{+}_{l,k,\sigma}(a_{l,k,\sigma})$ is the operator of creation (annihilation) for f- and d-electrons with 4-momentum k and spin quantum number σ $(=\pm 1)$; $N_{q,l} = \sum_\sigma N_{q,l,\sigma}$; $N_{q,l,\sigma} = \sum_{\mathbf{k}}a^{+}_{l,k,\sigma}a_{l,k+q,\sigma}$ is the Fourier transform of the operator of the density of electrons with spin σ at the site; k and q are quasimomenta; $\mathbf{S}_{q,l}$ is the Fourier transform of the operator of the vector spin density of electrons.

According to the spin-fluctuation theory of magnetic phase transitions in strongly correlated electron systems (see, e.g., [8,10]), the competition between the itinerant motion of electrons and the electron-electron interaction causes space-time fluctuations of the internal exchange field (ξ) and the charge field (η). In the neighborhood of the magnetic phase transition, we can approximate $r_c \gg r_e$ and $\tau_{fl} \gg \tau_e$, where $r_e(\sim a)$ and $\tau_e(\sim T_F^{-1})$ are the length and the time of the electronic jumps, respectively, $r_c(\sim aD^{1/2})$ and $\tau_{fl}(\sim T_C^{-1})$ are the correlation length and the fluctuation of spin density (FSD), respectively, a is the interatomic distance, D is the exchange enhancement factor ($D\sim U\chi$, χ - magnetic susceptibility of the electrons), and T_F is the Fermi temperature. In this approximation, the expression for the electron density of states is given by [8]

$$g_{l,\sigma}(\varepsilon) = \sum_{\alpha=\pm 1}(1+\alpha\sigma M_l/m_l)g_{0,l}(\varepsilon+\alpha U^{(l)}m_l+U^{(l)}n_l/2)/2, \tag{4}$$

where $m_l = (\sum_\gamma\langle m^2_{l,\gamma}\rangle + M^2_{l,\gamma})^{1/2}$ is the component $(\gamma = z,\perp)$ root mean square of the magnetic moment at the site (for more details see [8]); $M_{l,\gamma}$ is the γ-th component of the homogeneous magnetization, which is determined by the magnetic state equation

$$M_{l,\gamma}(D^{-1}_{l,\gamma}+2\kappa_l M^2_{l,\gamma}) = 0; \tag{5}$$

κ_l is the mode-mode coupling constant (see, e.g., [11]), $\langle m^2_{l,\gamma}\rangle$ are the amplitude of the longitudinal $(\gamma = z)$ and transverse $(\gamma = \perp)$ spin fluctuations in a system of the d- or f-electrons (l=f or d, respectively), which is determined from the internal temperature T,

$$D^{-1}_{l,\gamma} = 1 - \frac{m_l^2 - m^2_{l,\gamma}}{{m_l}^3}n^{(l)}_{ef} - \frac{m^2_{l,\gamma}}{{m_l}^2}U_l\tilde{g}_l(\mu,m_l) \tag{6}$$

is the inverse of the exchange enhancement factor of the longitudinal $(\gamma = z)$ and the transverse $(\gamma = \perp)$ magnetic susceptibilities;

$$n^{(l)}_{ef} = \sum_{\alpha=\pm 1}\alpha\int g_{0,l}(\varepsilon)f(\varepsilon-\mu-\alpha U^{(l)}m_l+U^{(l)}n_l/2)/2 \tag{7}$$

is the effective number of magnetic carriers; n_l is the number of filled d- or f-electron states that are defined by the position of the chemical potential, which is obtained from the electroneutrality condition

$$n = n_f + n_d = \int\sum_\sigma(g_{f,\sigma}(\varepsilon)+g_{d,\sigma}(\varepsilon))f(\varepsilon-\mu)d\varepsilon; \tag{8}$$

and

$$\tilde{g}_l(\mu,\xi_l) = 2\prod_{\alpha=\pm 1}g_0{}^{(l)}(\mu+\alpha U^{(l)}m_l+U^{(l)}n_l/2)/\sum_{\alpha=\pm 1}g_0{}^{(l)}(\mu+\alpha U^{(l)}m_l + U^{(l)}n_l/2) \tag{9}$$

is the reduced density of state.

These relations are valid for the description of bulk and single-crystal planar magnetic semiconductors (three-dimensional and two-dimensional cases). For the ultra-thin films, the quasi two-dimensional situation occurs, where the band motion of the electrons and the exchange interactions are realized in the plane, but the vector spins, the exchange fields and the magnetization are three-dimensional. The spin splitting of the electron energy and the changes in the occupation numbers of electron states depend on the internal temperature, which is determined from the environment temperature, the applied voltage and the current density through the sample. Therefore, the system of Eqs. (1) and (4)–(9) must be supplemented with the following expression for the current density of mobile charge carriers

$$j = \sigma E. \tag{10}$$

We apply a simple expression for the conductivity

$$\sigma = \frac{e^2}{2m^*}n(T_0,U)\tau, \tag{11}$$

where m^+ and τ are the effective mass and the mean free time density of mobile charge carriers, respectively, which are calculated according to the electroneutrality condition. Expression (7) depends on the applied voltage and the time. In the stationary case, when $d/dt = 0$ in (1), we obtain

$$n(T_0,U) \equiv n(T_0 + JUh/S\lambda) \tag{12}$$

The resulting system of Eqs. (1) and (4)–(11) allows us to describe the current-voltage characteristics in the neighborhood of the magnetic phase transformation from the ferromagnetic metallic state to the semiconducting paramagnetic state, considering the effects of self-heating. According to Eqs. (4)–(8), the electric field significantly changes the magnetization because the amplitude of the FSD defines the internal temperature of the sample. Consequently, this change creates a gap in the energy spectrum of the electrons and subsequently increases its width when the applied voltage increases. Eqs. (1) and (4)–(11) were analyzed, and the characteristic values of the voltage were numerically estimated for single-crystal $EuO_{1-\delta}$ films; the films were square sections, whose total area of the side surfaces is $S = 4Lh$. (L is the size of the sample along the direction of the current, and h is the size of the sample along the direction of the heat flow. The directions of the electric current and the heat flow are mutually perpendicular.) Based on the results [7], the density of states of the f-electrons is simulated by $g_{0,f}(\varepsilon) = (7/2\Delta)\theta(|\varepsilon|-\Delta)$ with half-width $\Delta = 0.3$ eV. The density

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of states of the conduction band is assumed to be constant and equal to 2/3 eV^{-1} over the entire range of energies required for the calculations (≈ 0.5 eV). The bottom of the conduction band was located at 1.1 eV [7] above the upper limit of the energy range of the 4f7-multiplet. To describe the metallic ground state $EuO_{1-\delta}$, the energy position of the chemical potential is chosen in the energy range that corresponds to the 4f7-multiplet. Thus, the localized f-states are partially filled, and there is a ferromagnetic ordering. In conditioned magnetic ordering, the spin splitting energies of f- and d-electrons cause their spectra to overlap and the electrons to redistribute between the f- and the d-states. As a result, at $T=0$ K, the conduction band is partially filled.

To analyze the electronic subsystem $EuO_{1-\delta}$ in an external electric field, we consider the effect of the splitting of the electron energy spectra when the internal exchange fields fluctuate. For a given temperature and a given field, the dependence of the spin fluctuation amplitude is described by the following expression [8]

$$\left\langle m_{d,\gamma}^2 \right\rangle = B_d T^2/[U^{(d)2} D_{d,\gamma}^{-1}(D_{d,\gamma}^{-1}+a_d)], \left\langle m_{f,\gamma}^2 \right\rangle = D_{f,\gamma} T/U^{(f)}$$

with the following parameters: $Bd=3/\pi U^{(d)}$, $a_d=0.1$, $U^{(f)}=4$ eV, and $U^{(d)}=0.8$ eV.

In the stationary case, the internal temperature of the sample with square cross-section determined the expression $-T=T_0+jUh^2/4L\lambda$. Based on the calculated surface temperature of the film $T0<40$ K, the Joule heating of the sample decreases the magnetization (Fig. 1), as demonstrated in the concept of "hot magnon". In addition, the Joule heating reduces the root mean square of the magnetic moment, which makes the electronic states appear with both spin directions (see (2)) at the identical energies and reduces the magnitude of the magnetic splitting energy of f- and d-states; the latter shifts them away from each other. Hence, the magnitude Hubbard shift changes because the electrons redistribute between the f- and the d-states. The redistribution of electrons (with an increase in the filling of the localized f-states) increased the electrical resistance and formed a negative feedback between the current and the voltage. The dependence of the electrical resistivity on the applied voltage is nonlinear, which formed the bistable state along the density current and manifested in the appearance of an N-shaped portion in the CVC (see Fig. 2 (a)). If the internal temperature is set to ~40 K, the energy gap between the f- and the d-states is restored, and its width increases with increasing temperature. The result is a semiconductor state with a temperature-dependent band gap.

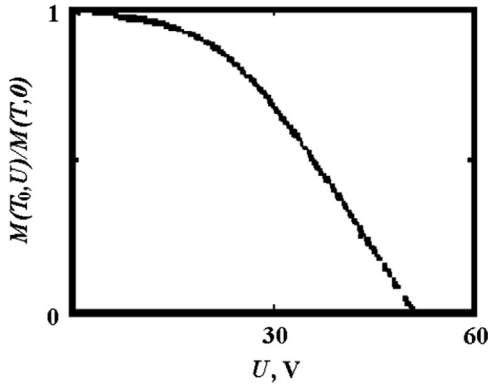

Fig. 1. Field dependence of the homogeneous magnetization of the $EuO_{1-\delta}$ film in the self-heating condition for $L/h=100$ at the film surface temperature of 30 K.

The numerical calculation results of the stationary current-voltage characteristics of the $EuO_{1-\delta}$ film for $T0>50$ K are shown in Fig. 2(b). According to the current-voltage characteristics, $EuO_{1-\delta}$ can have a non-equilibrium transition from the "cold" phase to the "hot" phase of the semiconductor. In this case, the analysis of the dependence of internal temperature on the applied voltage shows that the voltage transition in the "hot" phase corresponds to the required heat to raise the sample temperature to near 80 K, where the formation of the energy gap between the f- and the d-states stops and its width remains almost unchanged with further increase in internal temperature. With the achievement of 80 K, the internal temperature sharply increases to a determined external temperature and to the size of the sample.

During the reverse of the current-voltage characteristic, there is a transition in the "cold" phase. Initially, the internal temperature approaches TC. Because of the magnetic phase transition, the internal temperature of the sample abruptly decreases to a value less than TC.

In a bistable state of the electronic subsystem, Eq. (1) also has time-dependent solutions. In particular, they arise in the case of sufficiently thin films ($L/h>100$) and satisfy the self-oscillations of the current (S-shaped CVC) or the voltage (N-shaped CVC).

In the temperature range that corresponds to the N-shaped current-voltage characteristic (Fig. 2(b)), the attractor of the system of equations is the limit cycle, and the voltage only self-oscillates under a constant current density (see Fig. 3(a)). Subsequently, in the range of external temperatures that corresponds to the current, the bistability (Fig. 2(b)) is realized using a self-oscillation current at a constant external voltage (see Fig. 3(b)).

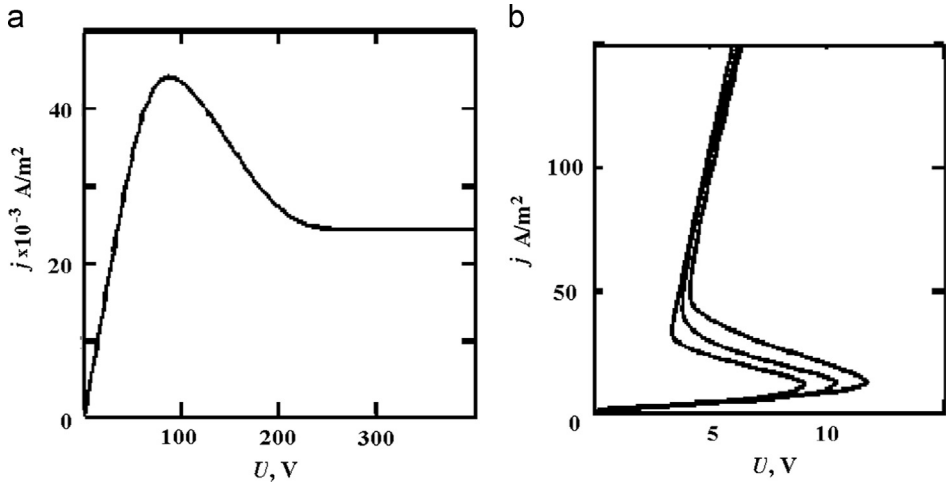

Fig. 2. Current-voltage characteristics of the film $EuO_{1-\delta}$ in the self-heating conditions for $L/h=100$ at the following film surface temperatures: (a) 30 K, (b) 55 K, 60 K and 65 K (from left to right).

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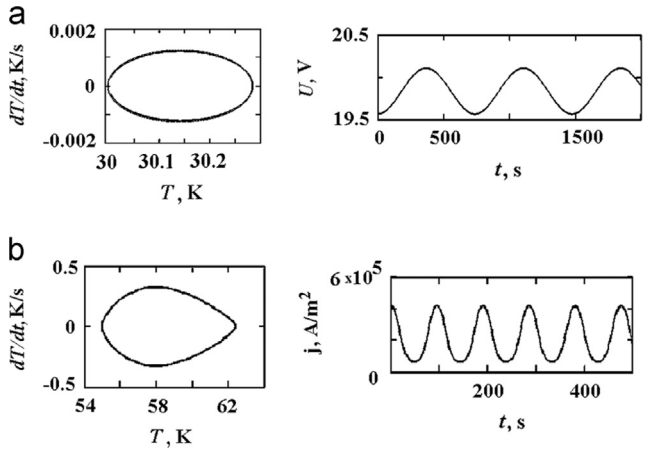

Fig. 3. Phase trajectory and periodic oscillations of the voltage (a) and the current density (b). $L/h=100$, $L=100$ mm; (a) $T_0=30$ K, $j=22$ mA/cm^2; (b) $T_0=55$ K, $U=50$ v.

Hysteresis effects that are associated with the self-heating of electron and magnetic subsystems are experimentally observed in $EuO_{1-\delta}$ films with a thickness of approximately 300 nm. Consistent with our analysis, the self-oscillation current or voltage should occur in thin-film samples. In particular, it is expected that in nanofilms, $EuO_{1-\delta}$ will only be the dynamic solutions ($dT/dt \neq 0$). The characteristic time of self-oscillation processes is determined from the heat transfer rate and significantly reduced in the transition to the nanoscale but remains much larger than the characteristic time of the spin fluctuations. Examples of these oscillations are shown in Fig. 3.

3. Conclusion

Thus, according to the spin-fluctuation theory of magnetic semiconductors, the electric field leads to the self-heating of both the electronic and the magnetic subsystems. This self-heating creates a bistability in the current or the voltage of the electrons, the magnons and the paramagnons. This process produces electrical-hysteresis effects in the voltage or current oscillations of the thin films.

Another group of substances for which these effects are also possible are rare earth manganites with a colossal magnetoresistance effect [12]. It is of interest to investigate the influence of electric-field self-heating on this effect.

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