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Magnetic contribution to electric resistance of gadolinium single crystal at low temperatures

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Electrical resistance of a single crystal of ferromagnetic rare-earth metal, gadolinium, has been measured within 150 to 325 K temperature range including the Curie point T_c and the spin reorientation temperature T_{sr}. The temperature dependences of magnetic contribution to the electric resistance, $(\rho/\rho)^{magn}$, along the main c axis and normal thereto are qualitatively different: along the c axis, this dependence has a convex curve shape while in the **ab** plane, it is a concave one showing a kink at T_{sr}.

Измерено электросопротивление монокристалла ферромагнитного редкоземельного металла гадолиния в диапазоне температур 150–325 К, включающим точку Кюри T_c и температуру спиновой переориентации T_{sr}. Установлено, что температурная зависимость магнитного вклада в электросопротивление $(\Delta\rho/\rho)^{magn}$ вдоль главной оси **c** и в перпендикулярном направлении носит качественно различный характер – выпуклая кривая вдоль оси **c** и вогнутая кривая с резким перегибом в T_{sr} для плоскости **ab**.

Gadolinium (Gd) occupies a specific place among heavy rare-earth metals (REM) for several reasons. First, Gd^{3+} ion having the $4f^75s^25p^6$ configuration has no orbital momentum while the spin momentum has the maximum known value ($S = 7/2$). Due to absence of spin-orbital interaction, the magnetic anisotropy of gadolinium is two orders of magnitude lower than those of other REMs with $L \neq 0$ (Dy, Yb, etc.). Second, among all REMs, it is Gd only that shows a characteristic direct paramagnetism-to-ferromagnetism transition ($P \rightarrow F$), the hexagonal crystal lattice symmetry being conserved in the ferromagnetic state, in contrast to other REMs (Dy, Tb, etc.). Note that the direct $P \rightarrow F$ transition was called in question in some earlier works [2, 3] although direct neutron diffraction studies [4] evidence the collinear ferromagnetic structure below the Curie point. Third, the Gd transition temperature to magnetically ordered state ($T_c \sim 293$ K) is the highest one among all REMs. Fourth, at temperature lowering down to $T_{sr} \sim 230$ K, Gd shows the spin relaxation transition from the high-temperature collinear ferromagnetic structure (the magnetic momentum **M** is parallel to the main **c** axis of hcp lattice) to the low-temperature conical one, the cone apex angle of the "disordered" conical structure 2φ (the main **c** axis being the cone one) being unmonotonically temperature-depended [4]: in contrast to the ferromagnetic FS spiral inherent, e.g., in Ho, the projections of Gd manetic moments on the basal plane do not form a regular spiral. Fifth, impurities effect appreciably T_c and T_{sr} [5].

Kinetic properties of single crystalline and polycrystalline gadolinium, first of all, its electric resistance, at low temperatures was studied in numerous works (see, e.g., [6–11]). In those works, attempts were made to discriminate the magnetic contribution to the resistance (ρ_{magn}) and to establish the character and nature of the

$\rho_{magn}(T)$ dependence. To that end, the Matthiessen rule [12] was used for not very low temperatures (when the contribution to the resistance due to electron-electron collisions, ρ_{el}, is negligible, the Debye temperature for Gd being rather low, Θ_D~180 K [13]). The rule is expressed as

$$\rho(T) = \rho_0 + \rho_{phon}(T) + \rho_{magn}(T), \quad (1)$$

where ρ_0 is the residual resistivity due to impurities and defects; ρ_{phon}, resistance due to scattering on phonons; ρ_{magn}, that due to scattering on magnons. Although ρ_{magn} was revealed in all the studies, the results obtained are contradictory. The contradictions concern questions of principal importance, namely, the $\rho_{magn}(T)$ dependence shape, the ρ_{magn} anisotropy character, the effects in magnetic phase transition(s) point(s).

In this connection, this work is aimed at precise study of electric resistance of a Gd single crystal in directions along and perpendicular to the main axis of hcp lattice within temperature range of 150 to 325 K including T_c and T_{sr}. The magnetic susceptibility was measured, too, to detect phase transitions in different directions. The ultimate purpose of the work is to establish a correlation between the behavior of magnetic contribution to gadolinium electric resistance and the magnetic structure evolution at temperature variation.

All studies were done using a gadolinium single crystal of about 2×2×10 mm^3 size obtained by zone recrystallization method. The material purity grade was at least 99.7 % (main impurities being REMs, amount of other impurities did not excess 0.03 %). The single crystal was oriented to within 1–2° using a special attachment to a DRON-4-17 X-ray diffractometer. The samples for measurements were cut using an electrical discharge machine. The current and potential contacts were applied using conductive silver adhesive.

The electric resistance and magnetic susceptibility of the Gd single crystal at low temperatures were studied using a PC-based instrument-software measuring complex (ISMC) [14]. The ISMC for measurements of temperature dependences of electrophysical and magnetic properties within the 10 to 300 K range has been developed using a RGD-210 cryogenerator (*Ley bold*). A platinum resistance thermometer was used as the temperature sensor.

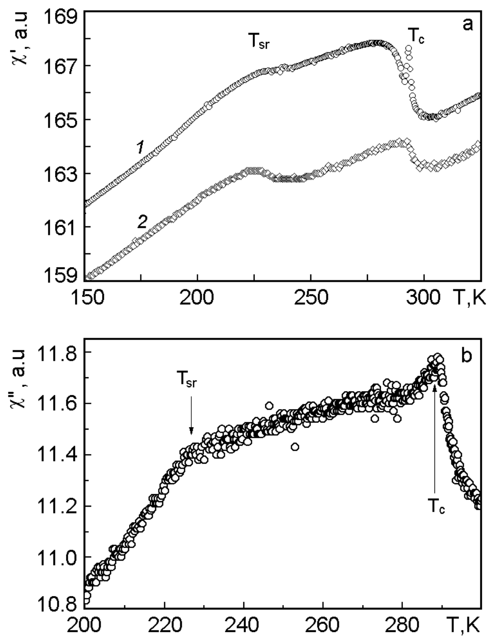

Fig. 1. Temperature dependence of real component of gadolinium complex magnetic susceptibility along (*1*) and perpendicular to the main axis (*2*) – (a). $\chi_c''(T)$ – (b).

As mentioned above, the dynamic magnetic susceptibility was measured as a function of temperature to detect the magnetic transformations at T_c and T_{sr}. The essence of the method consists in determination of changes in magnetic parameters of a coil due to introduction of the sample thereto [15]. To that end, the temperature dependences of inductance were determined for the sample-free coil [$L_{coil}(T)$], the coil with a sample [$L_{tot}(T)$], and the magnetic loss tangent [tgδ(T)] were determined using a E7-12 digital inductance meter (operating frequency f = 1 MHz). Since the measurements were rather qualitative and the $L_{coil}(T)$ is monotonous, it is possible to determine $L_{sample}(T) = L_{tot}(T) - L_{coil}(T)$ where L_{sample} is the contribution to the coil inductance due to the sample therein.

The values of real (χ') and imagine (χ'') components of the complex magnetic susceptibility

$$\chi = \chi' + i\chi'', \quad (2)$$

are related to L_x and tgδ by simple relationships

$$\chi' = A \cdot L_{sample}, \quad \chi'' = \chi' \cdot \mathrm{tg}\delta, \quad (3)$$

where A is the instrumental constant.

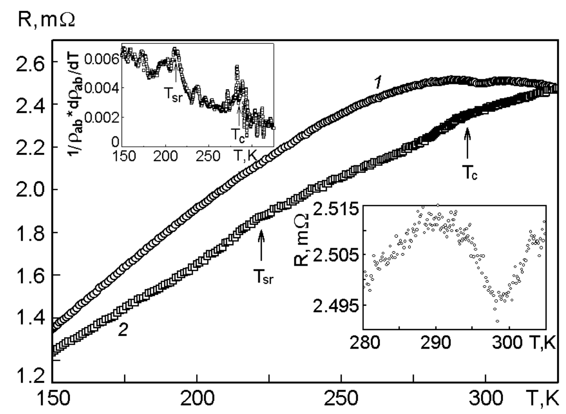

Fig. 2. Temperature dependence of gadolinium single crystal electric resistance along (*1*) and perpendicular to the main axis (*2*). Insets: $R_c(T)$ near T_c (right); temperature dependence of electric resistance temperature coefficient in the **ab** plane (left).

The measured values of quantities in proportion to the real (χ') and imagine (χ'') components of gadolinium magnetic susceptibility tensor along the main **c** axis and in the basal **ab** plane are presented in Fig. 1.

The temperature dependence of Gd magnetic susceptibility is seen to be anisotropic. At $T = T_c \sim 293$ K (the precise determination of magnetic phase transition temperatures is beyond the frame of this work), a pronounced peak is observed in the $\chi_c'(T)$ dependence that is typical of second order phase transitions; in $\chi_{ab}'(T)$ curves, the singularities in the Curie point are much less pronounced. In contrast, near the spin reorientation temperature $T_{sr} \sim 225$ K, the effects are seen to be a rather broad peak in the $\chi_{ab}'(T)$ curve and only a hint at a maximum in the $\chi_c'(T)$ one. The change in $\chi_c''(T)$ curve near T_{sr} is more clear, see inset in Fig. 1. The observed magnetic susceptibility anisotropy at 1 MHz agrees qualitatively with the low-frequency measurement results ($f = 250$ Hz [5]) while another anisotropy character of χ' and χ'' has been revealed in [3] at 10 to 1000 Hz frequencies. To discuss reasons for these distinctions is obviously beyond the frame of this work where measurements of $\chi(T)$ dependences play only auxiliary part as an indicative method.

The measurement results of electric resistance of the Gd single crystal along the main **c** axis and in the basal **ab** plane are presented in Fig. 2. Due to complicated shape of Gd single crystals, absolute (R) resistance values along the crystal axes are presented in the Figure instead the specific ones (ρ).

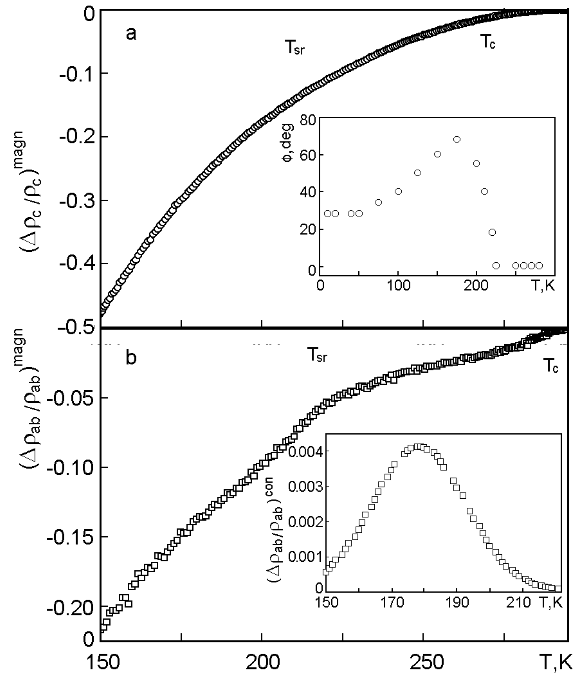

Fig. 3. Temperature dependence of magnetic contribution to gadolinium single crystal electric resistance along and perpendicular to the main axis. a) $(\Delta\rho_c/\rho_c)^{magn}(T)$, inset: temperature dependence of angle between the magnetic momentum **M** and **c** axis for gadolinium [4]. b) $(\Delta\rho_{ab}/\rho_{ab})^{magn}(T)$, inset: $(\Delta\rho_{ab}/\rho_{ab})^{con}(T)$.

The $R(T)$ curves along the **c** axis and perpendicular thereto are substantially different. At $T < T_c$, the $R_{ab}(T)$ curve is concave while the $R_c(T)$ one is convex. Two inflexions are observed in the $R_{ab}(T)$ dependence, one near T_c and second, near T_{sr}. These inflexions correspond to two maxima in the temperature dependence of resistance coefficient $(1/\rho_{ab})\cdot(d\rho_{ab}/dT)$, see *left-hand* inset in Fig. 2. In the $R_c(T)$ curve, there is only one maximum near T_c, see the *right-hand* inset in Fig. 2. No appreciable singularities are observed near T_{sr} in temperature dependences of R_c or $(1/\rho_c)\cdot(d\rho_c/dT)$.

To discriminate the magnetic component of gadolinium electric resistance, the contributions associated with electron scattering on phonons, R_{phon}, and the residual resistance R_0 are to be subtracted from the obtained $R(T)$ dependences. Since in this work, measurements were done at relatively high temperatures (150–325 K) and the Debye temperature for Gd is rather low, the

$R(T)$ dependence in the paramagnetic region can be approximated by a straight line

$$R(T) = R_0 + \frac{dR_{phon}}{dT} \cdot T = A + B \cdot T. \quad (4)$$

Subtracting this straight line from the experimental $R(T)$ curve, the magnetic contribution to the resistance can be obtained at a certain accuracy. Unfortunately, the magnetic contribution discrimination procedure has been applied successfully only to $R_{ab}(T)$ dependence. The magnetic contribution along the main axis can be discriminated only very approximately, since there is no linear section with positive slope in $R_c(T)$ curve at $293 \le T \le 325$ K.

The temperature dependence of relative magnetic contribution {for each experimental point (T, R) lying below T_c, $(\Delta\rho/\rho)^{magn} = [R(T) - A - BT]/R(T)$] to gadolinium electric resistance is shown in Fig. 3. Two distinct regions are observed in the $(\rho_{ab}/\rho_{ab})^{magn}(T)$ curve. Those are, a section of relatively weak almost linear temperature dependence ($T_{sr} < T < T_c$) and that of a higher slope and an appreciable nonlinearity ($T < T_{sr}$). The $(\Delta\rho_c/\rho_c)^{magn}(T)$ curve is parabolic within the whole $T < T_c$ range and no singularities are observed near T_{sr}.

Thus, the main results of electric resistance study of a gadolinium single crystal are as follows. In ferromagnetic state, gadolinium exhibits a strong resistance anisotropy. The $R_c(T)$ dependence is a curve with a maximum near the Curie point T_c, the $R_{ab}(T)$ one shows two inflections near the Curie point and the spin reorientation temperature T_{sr}. The temperature dependences of magnetic contribution to gadolinium electric resistance along the main axis and perpendicular thereto are different qualitatively. Along the **c** axis, the dependence is presented by a parabolic curve without any singularities at T_{sr}, in the **ab** plane, the curve has a kink at T_{sr}.

The latter instance (not noted up to date as far as we know) seems to be of considerable interest. Such a behavior of $(\Delta\rho/\rho)^{magn}(T)$ dependences results obviously from effect of gadolinium magnetic structure evolution on its kinetic properties. In fact, within the whole temperature range $T < T_c$, there is a component of magnetic momentum **M** ($M_c = M\cdot\cos\varphi$) along the **c** axis, thus, a magnetic contribution to the resistance is present within the same range, too. It is obvious that the decrease of the longitudinal **M** component from M_c for the collinear ferromagnetic structure (at $T_{sr} < T < T_c$) in the second order phase transition point (T_{sr}) down to $M\cdot\cos\varphi$ for the conical structure (at $T < T_{sr}$) is not accompanied by a substantial change in the $R_c(T)$ dependence.

It is another situation for directions perpendicular to the main axis. The component of magnetic momentum **M** for directions lying in the **ab** plane ($M_c = M\cdot\sin\varphi$) appears only at $T = T_{sr}$, whereas the magnetic contribution to electric resistance changes sharply. The appearance of nonlinear contribution to $(\Delta\rho_{ab}/\rho_{ab})^{magn}$ is very symptomatic. It evidences a pronounced correlation between the extreme character of temperature dependence of the conical ferromagnetic structure parameter, i.e., the angle φ between the magnetic momentum and **c** axis (see inset in Fig. 3a) and extreme behavior of the corresponding contribution to $(\Delta\rho_{ab}/\rho_{ab})^{magn}$, i.e., $(\Delta\rho_{ab}/\rho_{ab})^{con}$ (see inset in Fig. 3b). The rather small $(\Delta\rho_{ab}/\rho_{ab})^{con}$ value was estimated as the deviation extent of the $(\Delta\rho_{ab}/\rho_{ab})^{magn}$ pendence from the linear one at $T<T_{sr}$.

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Магнітний внесок в електричний опір монокристала гадолінію при низьких температурах

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Виміряно електроопір монокристала феромагнітного рідкісноземельного металу гадолінію у діапазоні температур 150–325 K, який включає точку Кюрі T_c та температуру спінової переорієнтації T_{sr}. Встановлено, що температурна залежність магнітного внеску у електроопір $(\Delta\rho/\rho)^{magn}$ вздовж головної вісі **c** та у перпендикулярному напряму має якісно різний характер – выпукла крива вздовж вісі **c** та угнута крива із різким перетином T_{sr} для площини **ab**.