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Anomalies of kinetic, magnetic and relaxation properties of dysprosium in the region of helicoidal antiferromagnetic structure

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To reveal macroscopic effects associated with phase transitions between the commensurate and incommensurate phases, temperature dependences of ohmic resistivity ρ, complex magnetic susceptibility of attenuation $\chi = \chi' + i\chi''$ and low-frequency internal friction parameters, namely, the logarithmic decrement Q^{-1} and resonance frequency f have been studied experimentally for dysprosium metal within the temperature range of helicoidal antiferromagnetic structure existence $T_{F-AF} < T < T_N$. Anomalies of kinetic (ρ), magnetic (χ) and lattice (Q^{-1}, f) properties of dysprosium have been first revealed at temperatures corresponding to the helicoidal magnetic structure period **q** to the crystal lattice period along the **c** axis ratio values of about 6, 5.5, and 5.

С целью обнаружения макроскопических эффектов, связанных с фазовыми переходами между соизмеримой и несоизмеримой фазами, экспериментально изучены температурные зависимости удельного омического электросопротивления ρ, комплексной магнитной восприимчивости $\chi = \chi' + i\chi''$ и параметров низкочастотного внутреннего трения — логарифмического декремента затухания Q^{-1} и резонансной частоты f — редкоземельного металла диспрозия в температурном диапазоне существования геликоидальной антиферромагнитной структуры $T_{F-AF} < T < T_N$. Впервые обнаружены аномалии кинетических (ρ), магнитных (χ) и решеточных (Q^{-1}, f) свойств диспрозия при температурах соответствующих отношениям периода геликоидальной магнитной структуры **q** к периоду кристаллической решетки вдоль оси геликоида **c**, равным ~6, ~5,5 и ~5.

The crystal, magnetic and electron structure and various physical properties of a 4f-magnetic, rare-earth metal dysprosium (Dy) are under study for more than 50 years (see, e.g., reviews [1–5]). It has been established reliably that at T_N about 180 K, dysprosium is subjected to the second order phase transition (paramagnetism-to-antiferromagnetism), its hcp crystal lattice symmetry remaining unchanged. Studies of the neutron magnetic scattering [6] and synchrotronic X-ray magnetic scattering [7] characterize unambiguously the character of the antiferromagnetic structure in Dy. Namely, below T_N, a helicoidal antiferromagnetic structure of the simple helix SS type is formed with the helicoid axis directed along the 6th order symmetry axis (the **c** axis of hcp lattice). The magnetic moments **M** are positioned of course in the basis (001) planes. As the temperature drops down to T_{F-AF} (about 90 K), the 1st order phase transition occurs in Dy (helicoidal antiferromagnetism-to-collinear ferromagnetism) while the hcp lattice symmetry is reduced to orthorhombic one [8–11], and magnetic moments in the ferromagnetic phase are positioned in the basis (001) planes along the **a** axis.

In the temperature range $T_{F-AF} < T < T_N$, the angle between the **M** projections in neighboring basis planes (the helicoid angle θ) varies within very wide limits. So at $T = T_N$, θ is about 43.2° while at $T = T_{F-AF}$, about 26.5°. Not long ago, when studying the neutron magnetic scattering on a Dy single crystal [12], it has been found that the temperature dependence of the helicoid angle $\theta(T)$ is not monotonous (as was postulated in [6]) but contains a series of peculiarities ("jumps"). These peculiarities is believed to be associated with the appearance of so-called "commensurability points", that is, with transformation of the SS-lattice (where the period ratio between the magnetic lattice, $\mathbf{q} = (\pi/\theta)\cdot\mathbf{c}$, and the crystal one, **c**, is an irrational number) into a magnetic lattice where the period **q** is a multiple of one or more **c** periods [13].

The concept of realizability of phase transitions between commensurate and incommensurate phases (CI transition) goes back to works by Ya.I.Frenkel and T.A.Kontorova [14]. Their simplest one-dimensional model was further successively applied to many systems, including helicoidal magnetic structures (see, e.g., [15, 16]). The CI transitions are considered theoretically in numerous works (see, e.g., [17–20]). It has been assumed, in particular, that the occurrence of CI transitions as well as helicoidal, non-collinear, and other magnetic structures in rate-earth metals could be due to changes in the Fermi surface topology at temperature lowering or under external magnetic field [21, 22].

The experimental results concerning the transitions between the commensurate and incommensurate phases in the existence range of antiferromagnetic structure in Dy are highly contradictory. For example, numerous anomalies are mentioned to be observed in the temperature dependences of thermal expansion coefficient, elastic constants, and heat capacity of Dy within the $T_{F-AF} < T < T_N$ range (see, e.g., [23, 24]). At the same time, there are data evidencing the avbsence of any peculiarities in those properties (see, e.g., [25, 26]). No anomalies have been revealed in temperature dependences of the crystal lattice parameters **a** and **c** as well as **c**/**a** ratio for Dy in the $T_{F-AF} < T < T_N$ range [8–10]. The existence of anomalies in macroscopic properties near the "commensurability points" was stated to be doubtful [27] as well as even the existence of the "commensurability points" in dysprosium [22].

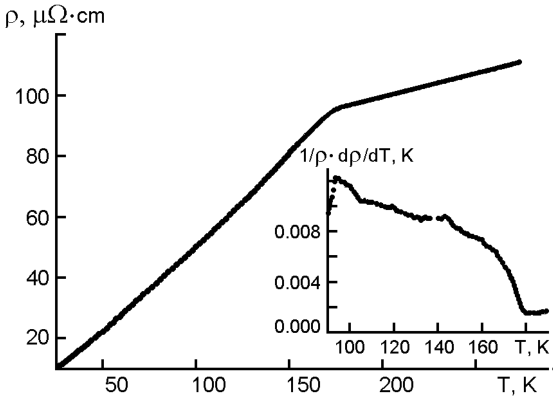

Fig. 1. Temperature dependence of dysprosium resistivity. Inset: temperature dependence of resistivity coefficient $1/\rho\ d\rho/dT$ at $T_{F-AF} < T < T_N$.

Due to a strong temperature dependence of the helicoidal antiferromagnetic structure period, **q**, as well as to presence of two reference points, namely, the pronounced magnetic phase transitions of 1st (at T_{F-AF}) and 2st order (at T_N), dysprosium can be used as a modeling object to consider the CI transitions in rare-earth metals. That is why in this work, we attempted to reveal anomalies in the electron and lattice properties of that metal at $T_{F-AF} < T < T_N$.

The investigation program included the measurements of temperature dependences of resistivity ρ, complex magnetic susceptibility $\chi = \chi' + i\chi''$ and low-frequency internal friction (IF) parameters, namely, the logarithmic decrement of attenuation Q^{-1} and resonance frequency f, in dysprosium at low temperatures (20–273 K for ρ and χ and 70–273 K for IF).

All the measurements were done on one and the same sample of 99.7 % pure polycrystalline dysprosium with residual resistivity $\mathrm{RRR} = \rho_{273\ \mathrm{K}}/\rho_{20.3\ \mathrm{K}} = 14$ shaped as a 18 mm long wire of 0.75 mm in diameter. The heating rate was about 0.1 K/s (for ρ and χ examinations) and about 1 K/s (for IF studies). The apparatus and procedure of those measurements have been described in [28–30].

Fig. 1 presents the temperature dependence of dc-resistivity (ρ) for dysprosium. A pronounced inflection is observed in the $\rho(T)$ curve near $T_{F-AF} = 86$ K. The weak sensitivity of resistivity to the F–AF phase transition is known to be associated with the fact that near T_{F-AF} and T_{AF-F}, the magnetically heterogeneous states (coexistence of ferro- and antiferromagnetic phases) are realized [10, 31]. No appreciable

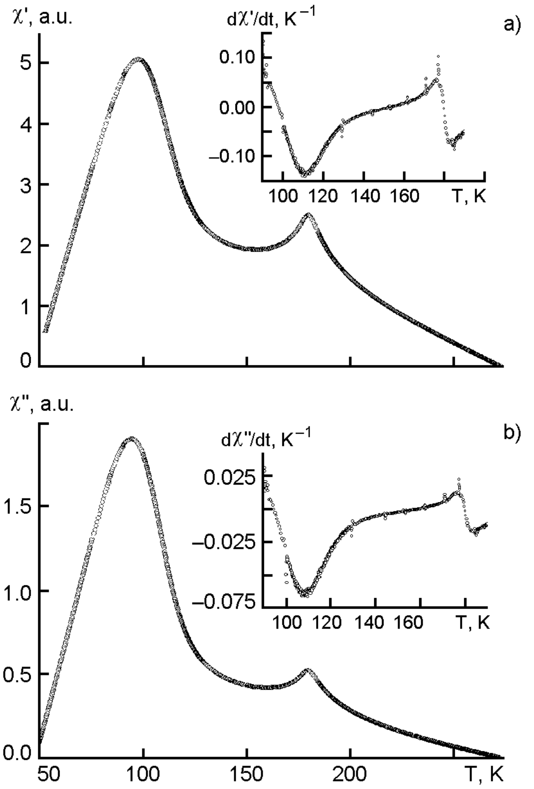

Fig. 2. Temperature dependence of real (a) and imaginary (b) parts of complex magnetic susceptibility for dysprosium. Insets: temperature dependences of $d\chi'/dT$ and $d\chi''/dT$ derivatives at $T_{F\text{-}AF} < T < T_N$.

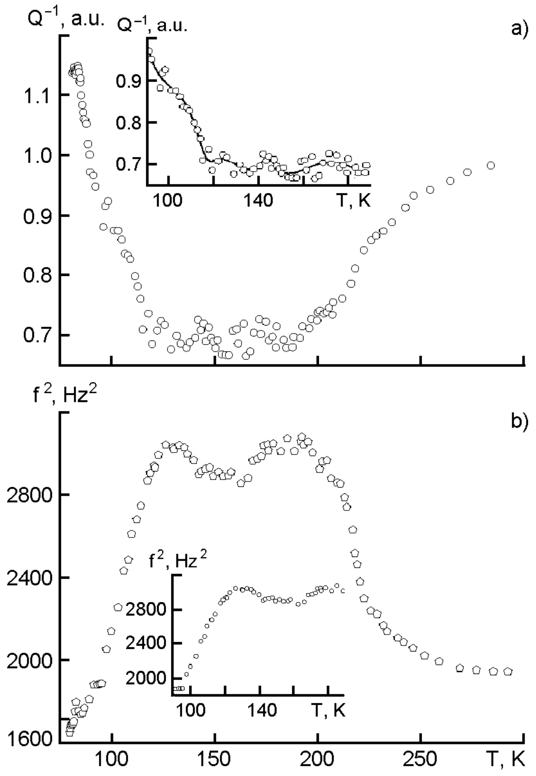

Fig. 3. Temperature dependence of internal friction parameters for dysprosium: logarithmic decrement Q^{-1} (a) and squared resonance frequency f^2 (b). Insets: $Q^{-1}(T)$ and $f^2(T)$ dependences at $T_{F\text{-}AF} < T < T_N$.

peculiarities have been revealed in the $T_{F\text{-}AF} < T < T_N$ range. However, the temperature dependence of the resistivity logarithmic derivative (inset in Fig. 1), besides of the anomaly characteristic of the 2nd order phase transitions at T_N (see, e.g., [32]), shows several effects, namely, a rather sharp slope change of $1/\rho(d\rho/dT)$ curve at $T \sim 107$ K and small but pronounced maxima at $T \sim 123$ K and ~140 K.

The temperature dependences of real and imaginary parts of high-frequency ($f = 1$ MHz) complex magnetic susceptibility in the $T_{F\text{-}AF} < T < T_N$ range are presented in Fig. 2. The $\chi'(T)$ and $i\chi''(T)$ curves show maxima near T_N. It is to note that, according to the Landau theory [33], the magnetic susceptibility of an antiferromagnetic is continuous and finite at T_N while its derivative $d\chi/dT$ makes a jump. The presence of a 1st order magnetic phase transition at $T_{F\text{-}AF}$ does not influence substantially the temperature dependences of χ' and $i\chi''$. The "giant" maxima of real and imaginary parts of magnetic susceptibility at $T \sim 95$ K are observed, too. The $d\chi'/dT(T)$ and $d\chi''/dT(T)$ curves show at T_N characteristic jumps predicted by the Landau theory [33]. Besides, rather wide negative anomalies centered at T ~107 K are present. No other peculiarities exceeding the measurement errors have been revealed in $d\chi'/dT(T)$ and $d\chi''/dT(T)$ dependences.

The temperature dependences of the internal friction parameters (Q^{-1} and f^2) are presented in Fig. 3. The $Q^{-1}(T)$ curves (Fig. 3a) exhibit several rather well pronounced inflections at $T \sim 107$, 123 and ~140 K, besides of peculiarities characteristic of the 2nd (T_N) and 1st ($T_{F\text{-}AF}$) order phase transitions (Q^{-1} peaks). Although the relaxation measurements were done at a relatively low precision, the observed peculiarities of $Q^{-1}(T)$ dependence were reproduced well in all the measurement series carried out. The temperature dependence of the squared resonance frequency f^2 (Fig. 3b), that is, of the quantity proportional to the 2nd derivative of thermodynamic potential with respect to pressure (elastic modulus E), is pronouncedly anomalous at $T < T_N$. Namely, as the temperature rises from

T_{F-AF} up to about 123 K, f^2 increases; between 123 K and T_N, a crevasse is observed in $f^2(T)$ curve. Some anomalies in Q^{-1} and f^2 behavior were observed before at $T < T_N$ when measuring the high-frequency IF [34, 35]. Moreover, the $f^2(T)$ dependence shows trends to jumps in f^2 at 107 and 140 K, although the corresponding effects are very weak against rather large experimental errors.

Thus, in this work, anomalies of kinetic (ρ), magnetic (χ) and lattice (Q^{-1}, f) properties of dysprosium have been revealed within the temperature range of helicoidal antiferromagnetic structure existence. The strongest effect is the appearance of the magnetic susceptibility maximum and of $d\chi'/dT(T)$ and $d\chi''/dT(T)$ minima at $T \sim 107$ K. It is to note that the temperature corresponds to the helicoid angle $\theta = 30°$, that is, to $\mathbf{q}/\mathbf{c}$ ratio 6 [6]. Before, a magnetic anisotropy was revealed in Dy in the basic plane [36]. Moreover, less pronounced anomalies have been observed at the magnetic helix period ($\mathbf{q}$) multiples to 5.5 ($T_{5.5} \sim 123$ K) and 5 ($T_5 \sim 140$ K) periods of the crystal lattice ($\mathbf{c}$).

The presence of those anomalies at $T_{F-AF} < T < T_N$ could be considered as a consequence of certain peculiarities in the temperature dependence of the magnetic helicoidal structure period $\mathbf{q}$ (or of the helicoid wave vector $\mathbf{k}_H = (\pi/\theta)\cdot\mathbf{c}^*$ where $\mathbf{c}^*$ is the reciprocal lattice period) or, in other words, as an experimental confirmation to the existence of CI transitions in dysprosium.

While confirming the real existence of CI transitions in dysprosium, the results obtained do not explain uniquely the nature thereof. The behavior similarity of magnetic susceptibility, resistivity, and logarithmic decrement of attenuation at T_N and at $T \sim 107$ K allows to assume that the six-layer helicoidal structure formation at $T_6 \sim 107$ K is a 2nd order phase transition. As to relatively weak effects at $T_{5.5} \sim 123$ K and $T_5 \sim 140$ K, those could be only supposed to be associated with the topology changes of Fermi surfaces [21, 22, 37], that is, with electron topologic phase transitions of 2.5 order (in terms by Lifshits [38]). It is obvious that further experimental and theoretical studies are required to explain definitely the nature of CI transitions in rare-earth helicoidal magnetics.

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Аномалії кінетичних, магнітних і релаксаційних властивостей рідкісноземельного металу диспрозію в області існування гелікоїдальної антиферомагнітної структури

В.М.Аржавітін, В.В.Дерев'янко, Т.В.Сухарева, В.О.Фінкель

З метою виявлення макроскопічних ефектів, пов'язаних з фазовими переходами між співмірною і неспівмірною фазами, експериментально вивчено температурні залежності питомого омічного електроопору ρ, комплексної магнітної сприйнятливості $\chi = \chi' + i\chi''$ й параметрів низькочастотного внутрішнього тертя — логарифмічного декременту загасання Q^{-1} і резонансної частоти f — рідкісноземельного металу диспрозію у температурному діапазоні існування гелікоїдальної антиферомагнітної структури $T_{F-AF} < T < T_N$. Уперше виявлено аномалії кінетичних (ρ), магнітних (χ) і граткових (Q^{-1}, f) властивостей диспрозію при температурах, відповідних відносинам періоду гелікоїдальної магнітної структури **q** до періоду кристалічної гратки уздовж осі гелікоїда **c**, рівним ~6, ~5,5 і ~5.