Magnetic properties of a $GdCu_2$ single crystal

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The magnetization and magnetoelastic properties of single crystals and polycrystals of the compound $GdCu_2$ are investigated in fields up to 150 kOe. It is shown that in $GdCu_2$ an external magnetic field induces metamagnetic transitions of the spin-flip type. In order to interpret the observed features of the magnetization curves the energies of a series of magnetic configurations permitted by the symmetry of $GdCu_2$ are calculated and the possible changes of the magnetic structure in an external field are considered.

INTRODUCTION

The intermetallic compounds RCu_2 (where R is a rare-earth element or yttrium) have an orthorhombic crystal structure of the $CeCu_2$ type (space group *Imma*) (Ref. 1). The investigations of Refs. 2 and 3, performed on single-crystal and polycrystalline samples, have shown that with magnetic rare earths (RE) these compounds are antiferromagnetic at low temperatures, and many of them display metamagnetic behavior in an external magnetic field.[2,3] Neutron-scattering investigations show that the compounds RCu_2 have complicated magnetic structures that are often incommensurate with the crystal lattice.[4–6]

The interest in the compounds RCu_2 is connected with the fact that they are a good model object for verifying the possibilities of applying the Ruderman-Kittel-Kasuya-Yosida (RKKY) theory of exchange interaction via conduction electrons to RE intermetallic compounds. It has been shown[3,7,8] that in the framework of the RKKY model the change of the magnetic properties of the solid solutions $R(Cu_{1-x}Ni_x)_2$, where R = Gd or Tb, and, in particular, the change from antiferromagnetic to ferromagnetic ordering when the nickel content increases to 0.13 can be explained by the change of the concentration of conduction electrons and by the value (determined by this concentration) of the Fermi quasimomentum k_F

It is necessary to note that the application of the RKKY theory to the analysis of the properties of RCu_2 compounds with nonzero orbital angular momentum of the ion R^{3+} is complicated by the need to make correct allowance for the single-ion anisotropy energy, the magnitude of which in RCu_2 is comparable to the magnitude of the isotropic-exchange energy.

The most convenient object for the study of RKKY exchange in the series RCu_2 is $GdCu_2$, since the orbital angular momentum of the Gd^{3+} ions is equal to zero and the single-ion magnetic anisotropy of this compound is small. However, $GdCu_2$ has been studied considerably less than other RCu_2 compounds. This is due to the fact that, first, because of the large cross section for scattering of neutrons by gadolinium it has not been possible to perform neutron-scattering investigations of the magnetic structure of $GdCu_2$, and, second, measurements of the magnetic properties of the compound $GdCu_2$ have been performed only on polycrystalline samples in fields up to 60 kOe, which are insufficient for the destruction of the antiferromagnetic structure (although even in these fields phenomena that can be interpreted as the onset of a metamagnetic transition are observed[2,7]).

In this paper we describe investigations of the magnetization and susceptibility, and also of the magnetoelastic properties of single crystals and polycrystals of the compound $GdCu_2$ in strong magnetic fields up to 150 kOe. To interpret the observed features of the magnetization curves in the RKKY model we calculate the energies of a number of magnetic configurations permitted by the symmetry of the group Imma and determine the possible rearrangement paths of the $GdCu_2$ magnetic structure in an external field.

2. THE SAMPLES AND EXPERIMENTAL METHOD

Polycrystalline samples of $CdCu_2$ were fused in an electric-arc furnace on the cold hearth in an argon atmosphere and then homogenized at a temperature of 750 °C in a vacuum. The single-phase nature of the samples was monitored by the x-ray method.

The $GdCu_2$ single crystal was grown in a resistance oven with a temperature gradient of 10 °C/cm along the vertical in an argon atmosphere. A polycrystalline ingot of mass ~40 g was heated in a crucible of Al_2O_3 to a temperature (900 °C) exceeding the melting point of $GdCu_2$ (860 °C), and was held at this temperature for four hours. The sample was cooled at a rate of approximately 50 °C/h. A single-crystal sample of mass 35 mg was cut from the lower part of the ingot obtained. The lattice constants of the single crystal at room temperature ($a = 4.325$ Å, $b = 6.877$ Å, $c = 7.323$ Å) were close to the lattice constants of the polycrystal.

The magnetization was measured in the temperature range 4.2–270 K in static fields up to 60 kOe, and also in pulsed fields up to 150 kOe. The measurements in static fields were performed by the vibration-magnetometer method, with an error of less than 3%. In the measurements in pulsed fields an induction technique was used, and the error of the measurements of the absolute magnitude of the magnetization was less than 7–8%.

In the temperature range 7–300 K the lattice constants of $GdCu_2$ were investigated by the x-ray method. The measurements were performed on powders using the x-ray reflections (200), (031), and (103), and on the single crystal using the reflections (400), (060), and (006); Cu–K_α radiation was used. The relative accuracy of the measurements

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of the change of the lattice parameters with temperature for the single crystal was $\pm 1 \cdot 10^{-4}$ A, which was better than the relative accuracy for the powder by a factor of about 2.

The Young's modulus E of the polycrystalline $GdCu_2$ sample was measured by the combined-vibrator method[9] in the temperature range 4.2–300 K at a frequency of 200 kHz. The relative error of the measurements of the Young modulus as a function of the temperature was less than 0.03%.

3. EXPERIMENTAL DATA

In the paramagnetic region of temperatures above 50 K the magnetic susceptibility of the $GdCu_2$ single crystal does not depend on the direction of the external field and obeys the Curie-Weiss law with a paramagnetic Curie temperature $\theta_p = 16 \pm 2$ K and an effective magnetic moment $\mu_{\text{eff}} = (8.14 \pm 0.2)\ \mu_B$ close to the theoretical value for the Gd^{3+} ion ($7.94\,\mu_B$) (Fig. 1). The magnetization measured in a field of 1.5 kOe experiences a discontinuity at 42 K, which is evidence of the transition to the antiferromagnetic state. This is manifested even more clearly in the measurement of the susceptibility of $GdCu_2$ in a weak (~ 10 Oe) oscillating field (the insert in Fig. 1). The value of T_N for the single crystal agrees well with data in the literature[2] for a $GdCu_2$ polycrystal.

The transition to the antiferromagnetic state is also accompanied by magnetoelastic anomalies of the lattice parameters (Fig. 2) and of the Young's modulus (Fig. 3). It can be seen from Fig. 2 that below the Néel temperature T_N the lattice constants b and c of the orthorhombic unit cell increase and the lattice constant a decreases in comparison with the lattice constants obtained by extrapolation from the paramagnetic region of temperatures using the Debye law (the latter constants are shown in Fig. 2 by dashed curves). The magnetoelastic change of volume of the unit cell ($\Delta V/V = \Delta a/a + \Delta b/b + \Delta c/c$) is positive, and at 7.5 K reaches $(1.5 \pm 0.3) \cdot 10^{-3}$. In Refs. 10 and 11 the thermal expansion of a $GdCu_2$ polycrystal was measured by a dilatometric method and it was found that, at 4.2 K, $\Delta V/V = -0.96 \cdot 10^{-3}$, i.e., the magnetoelastic volume anomaly found in this work has the opposite sign. A possible reason for this contradiction may be the presence of crystalline texture with predominant orientation of the a axes of the crystallites along the direction of measurement of the thermal expansion of the polycrystalline samples investigated in Refs. 10 and 11. In this case, since $\Delta a/a < 0$ (see Fig. 2), the measured value of $\Delta V/V$ will also be negative.

The Young's modulus of $GdCu_2$ (Fig. 3) decreases near the Néel temperature, and upon further lowering of the temperature increases again. This nonmonotonic variation of the Young modulus was observed previously on polycrystalline samples of RE metals with a spiral magnetic structure.[12] The nonmonotonic dependence $E(T)$ is due to the competition of two mechanisms: The decrease of the modulus near T_N is due to the influence of elastic stresses on the magnetic structure, and the positive anomaly of the elas-

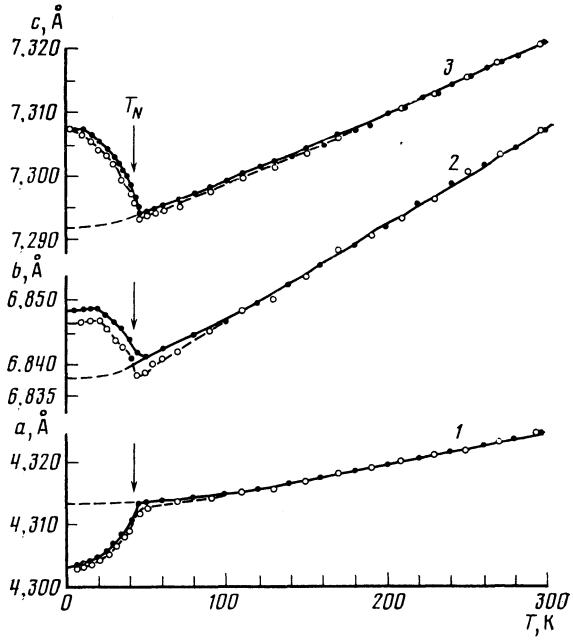

FIG. 2. Temperature dependences of the lattice constants of a $GdCu_2$ polycrystal (○) and single crystal (●), measured along the three principal crystallographic axes **a** (1), **b** (2), and **c** (3).

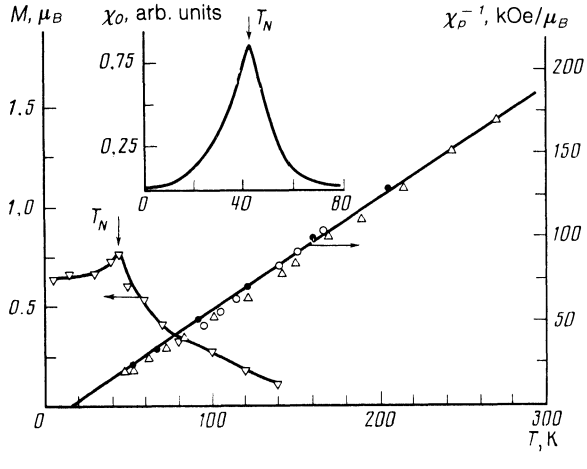

FIG. 1. Temperature dependence of the inverse paramagnetic susceptibility χ_p^{-1} of a $GdCu_2$ single crystal along the axes **a** (○), **b** (△), and **c** (●), and of the magnetization along the axis **c** (▽) in a field of 15 kOe. The temperature dependence of the initial susceptibility (in a field ~ 10 Oe) is shown in the insert.

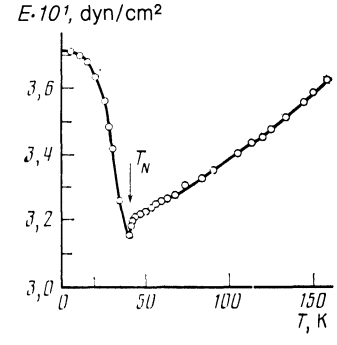

FIG. 3. Temperature dependence of the Young's modulus of $GdCu_2$.

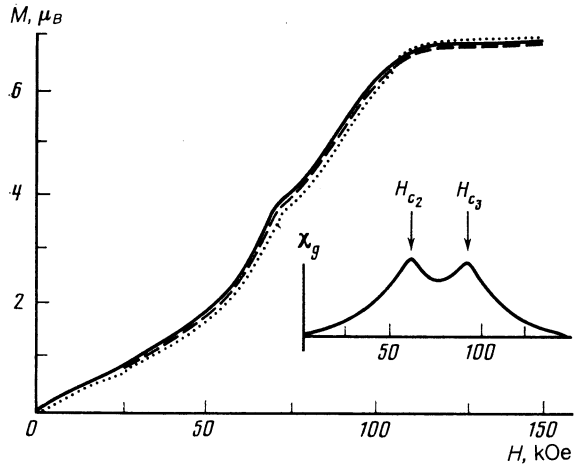

FIG. 4. Dependences $M(H)$ for the $GdCu_2$ single crystal, measured along the axes **a** (the solid line), **b** (the dashed line), and **c** (the dotted line) at 4.2 K. The insert shows the field dependence of the differential magnetic susceptibility χ_d along the axis **a**.

tic modulus at lower temperatures is due to the increase of the rigidity of the crystal lattice in the magnetically ordered state (for more detail, see Ref. 12).

The most interesting data were obtained below the Néel point in an investigation of the magnetization of $GdCu_2$ in strong magnetic fields. Figure 4 shows the magnetization along the three axes of the $GdCu_2$ single crystal at 4.2 K, measured in pulsed magnetic fields. In weak fields (up to 60 kOe) the magnetization depends linearly on the field, while upon further increase of the field a sharper increase of the magnetization is observed. In this case the saturation magnetic moment is equal to $(6.9 \pm 0.1)\,\mu_B$, which agrees well with the Gd^{3+} moment $(7.0\,\mu_B)$. This indicates that, in fields $H > H_{c3}$, $GdCu_2$ is in the ferromagnetic state. Thus, in the interval 60–120 kOe a metamagnetic transition from antiferromagnetism to ferromagnetism occurs in $GdCu_2$. A characteristic feature of this transition is the fact that it occurs not smoothly but, as it were, in two stages: At $H_{c2} = 68$ kOe the magnetization increases sharply, reaches approximately $4\,\mu_B$, and then increases more slowly; at $H_{c3} = 95$ kOe a sharp increase of the magnetization is observed again. The nonmonotonic character of the dependence $M(H)$ is clearly visible in the insert in Fig. 4, which shows the field dependence of the differential magnetic susceptibility of $GdCu_2$ at 4.2 K: The differential susceptibility passes through a maximum at the field values H_{c2} and H_{c3}.

With the field oriented along the **a** and **c** axes of the crystal a further small maximum of the susceptibility was observed in weak fields ~10–20 kOe (this is not shown in the insert in Fig. 4). This anomaly was studied in more detail in measurements of the magnetization in constant fields. Figure 5 shows magnetization curves for $GdCu_2$ at different temperatures in comparatively weak fields up to 28 kOe. It can be seen that in this range of fields the magnetization along the **b** axis increases linearly with the field, and the susceptibility does not depend on the temperature and is equal to $\chi_b = 4.1 \cdot 10^{-2}\,\mu_B$/kOe. The magnetization in weak fields along the **a** and **c** axes also depends linearly on the field, but is somewhat smaller than along the **b** axis $(\chi_a = \chi_c = 3.1 \cdot 10^{-2}\,\mu_B$/kOe). At a certain field value H_{c1} the magnetization displays a discontinuity that becomes sharper as the temperature increases; in this case, in the vicinity of the Néel temperature, the magnetization in the region of H_{c1} displays hysteresis. Above H_{c1} the susceptibilities χ_a and χ_c are equal to the susceptibility χ_b. Figure 6 shows the temperature dependences of the fields H_{c1}, H_{c2}, and H_{c3}. The fields of the magnetic transitions depend weakly on the temperature, and the transitions themselves are smeared out only in the immediate vicinity of T_N.

4. DISCUSSION OF THE RESULTS

Thus, from the experimental data obtained it follows that the compound $GdCu_2$ is a metamagnet: In this compound a transition from the antiferromagnetic to the ferromagnetic state occurs in a field. A characteristic feature in need of explanation is the fact that the destruction of the antiferromagnetic structure in a field occurs in two stages, in fields H_{c2} and H_{c3}.

Usually, a metamagnetic transition in antiferromagnets occurs in those cases when the magnetic-anisotropy energy is comparable to or larger than the isotropic-exchange energy.[13] For example, the metamagnetism in the compounds RCu_2 (R = Dy, Tb, etc.) is described in a 12-sublattice Ising model[14]; it is then possible, by choosing the exchange parameters, to explain the two-step metamagnetic transition observed experimentally in $DyCu_2$.

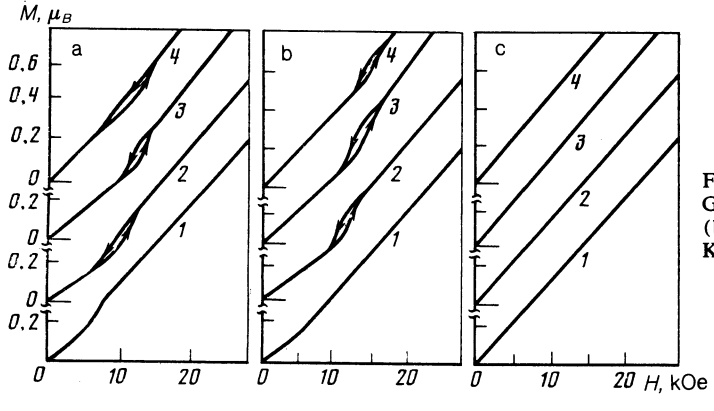

FIG. 5. Magnetization curves $M(H)$ of the intermetallic compound $GdCu_2$, measured along the three principal crystallographic axes **a** (a), **c** (b), and **b** (c) at different temperatures: 1) 4.2 K; 2) 15 K; 3) 30 K; 4) 35 K.

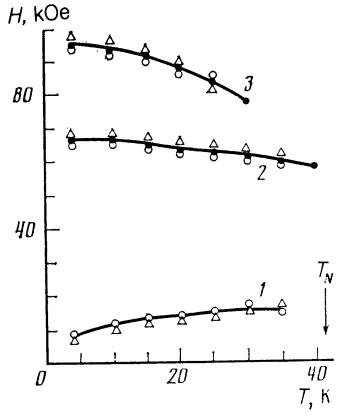

FIG. 6. Temperature dependences of the critical fields H_{c1} (1), H_{c2} (2), and H_{c3} (3) of the compound $GdCu_2$ along the three principal crystallographic axes: **a** (△), **b** (●), **c** (○).

Naturally, such a model is not appropriate for the description of the properties of $GdCu_2$ (although the metamagnetic transitions in $DyCu_2$ and $GdCu_2$ are outwardly similar), since the single-ion magnetic anisotropy of Gd^{3+}, as already noted, is small and the magnetic properties of $GdCu_2$ are practically isotropic. Therefore, in the first approximation it is necessary to treat the metamagnetism of $GdCu_2$ in an isotropic exchange model.

Poldy and Kirchmayr[3] have calculated the magnetic structure of $GdCu_2$ in the RKKY model. According to their calculations, in $GdCu_2$ spiral magnetic ordering occurs: The magnetic moments of the Gd^{3+} ions in each bc plane are ordered ferromagnetically, while the directions of the magnetic moments in neighboring planes differ by 35°.

With the aim of analyzing the possible changes of the magnetic structure of $GdCu_2$ compounds in an external magnetic field we have calculated the energies of the different magnetic phases permitted by the symmetry of the RCu_2 compounds in the RKKY-exchange model. Here, in contrast to Ref. 3, we have assumed that the mean free path of the conduction electrons is infinite. In this case the calculation of the energies of the different magnetic configurations reduces to the determination of lattice sums of the form

$$\sum_{i\neq j} F(2k_F R_{ij}) \exp(i\mathbf{Q}\mathbf{R}_{ij}), \tag{1}$$

$$F(x) = (x\cos x - \sin x)/x^4, \tag{2}$$

where $F(x)$ is the RKKY function,[15-17] k_F is the Fermi quasimomentum, and **Q** is the wave vector of the magnetic structure. The summation in (2) runs over all lattice sites over spheres of increasing radius, and is truncated after a given accuracy is reached. To accelerate the convergence one can use the method of Ref. 18, which is analogous to the Ewald method employed for the calculation of the energies of the electrostatic interactions in crystals. In the analysis of the specific magnetic structures in the compound $GdCu_2$ it is convenient to relabel the different crystallographically inequivalent positions of the Gd^{3+} ions in the unit cell as follows: 1 is (0, 0, 0), 2 is (0, 1/4, z), 3 is (0, 3/4, 1 − z), and 4 is ($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$), where $z = 0.5377$ (Ref. 1). It is not difficult to see that the Gd^{3+} ions form periodically alternating layers, lying in the *bc* plane, and the sublattices 1, 2, 3, and 4 belong to neighboring layers.

In Fig. 7 we give the calculated dependences of the energies of the ferromagnetic, ferrimagnetic, and spiral (investigated in Ref. 3) structures on k_F. As can be seen from the figure, the latter structure indeed possesses the lowest energy in the range of values $k_F = 1.349$–1.564 Å^{-1}. In Ref. 3, however, the orientation of the spins of the Gd^{3+} ions was not specified, since the energy of the isotropic RKKY exchange depends only on the relative orientation of the spins and does not change when they are rotated simultaneously.

In order to determine the orientation of the spins relative to the crystal axes, we turn to the experimental data. As can be seen from Fig. 5, the curves of the magnetization of $GdCu_2$ along the **a** and **c** axes practically coincide with each other, but differ from the curve of the magnetization along the **b** axis. But after the rearrangement of the magnetic structure in the critical field H_{c1} the slope of the curves $M_{a,c}(H)$ coincides with the slope of the curve $M_b(H)$. In other words, the differential susceptibilities χ_a, χ_c, and χ_b for $H > H_{c1}$ coincide. The experimental results obtained can be explained by assuming that for $H < H_{c1}$ the spins of the Gd^{3+} ions lie in the *ac* plane. [Here, of course, the angle between the spins of neighboring *bc* layers is 35°, and the wave vector of the structure is $\mathbf{Q} = (7\pi/18a, 0, 0)$ (Ref. 3)]. But the application of a field along the **a** or **c** axis leads to a simultaneous reorientation of all the spins into the *bc* or *ab* plane, respectively; i.e., a spin-flop transition occurs. In order that such a rearrangement of the magnetic structure in the field be possible, it is necessary that two conditions be fulfilled: First, the susceptibility $\chi_\perp$ of the system of spins lying in planes perpendicular to the magnetic field should be greater than the susceptibility $\chi_\parallel$ in the case when the spins are oriented in the ac plane; second, the gain in Zeeman energy of the system when the spins rotate into the *bc* or *ab* plane should exceed the loss in magnetic-anisotropy energy; i.e., the magnetic field should be greater than a certain

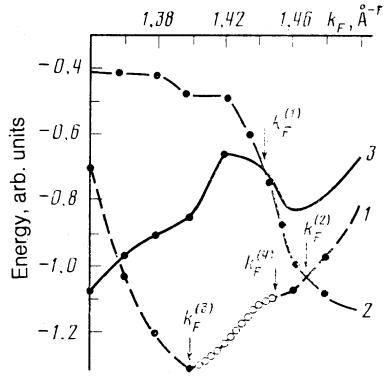

FIG. 7. Dependence of the energies of the spiral (1), ferrimagnetic [determined by (3)] (2), and ferromagnetic (3) structures on the quantity k_F. In the interval $k_F^{(3)} < k_F \leqslant k_F^{(4)}$ for $H > H_{c1}$ a rotation of the spirally ordered spins into the plane perpendicular to the external magnetic field **H** is possible.

critical value: $H > H_{c1} = [\Delta E_{an}/(\chi_\perp - \chi_\parallel)]^{1/2}$, where $\Delta E_{an} = E_{an}^{(ab)(bc)} - E_{an}^{ac}$ is the difference of the magnetic-anisotropy energies for orientation of the Gd^{3+} spins in the planes *ab* (*bc*) and *ac*. Because of the small magnitude of ΔE_{an} for the Gd^{3+} ions the field H_{c1} should also be small. In fact, as we see from Fig. 5, it satisfies $H_{c1} = 10$ kOe.

The features of the magnetization curve that are observable in the range 50–100 kOe can be related naturally to the appearance of an intermediate ferrimagnetic structure. In fact, our calculations show that in the range $k_F^{(1)} = 1.442$ Å$^{-1} \leqslant k_F \leqslant k_F^{(2)} = 1.464$ Å^{-1} the energy of the ferrimagnetic structure, for which the spins of the sublattices 1–4 are determined by the relations

$$\mathbf{S}_{2n}=\mathbf{S}_{3n}=\mathbf{S}_{4n}=\mathbf{S}_0, \quad \mathbf{S}_{1n}=\frac{4}{3}\mathbf{S}_0\left[-\frac{1}{4}+\cos\left(\frac{2\pi}{3a}\mathbf{q}_0\mathbf{R}_{1n}\right)\right], \tag{3}$$

lies between the energies of the spiral phase and ferromagnetic phase (see Fig. 7). In (3), $\mathbf{q}_0 = (1, 0, 0)$ and n labels the different unit cells.

For a more detailed investigation of the behavior of $GdCu_2$ in an external magnetic field **H** from the conditions for the minimum of the total energy, including the RKKY exchange and the Zeeman term, the equilibrium orientations of the spins for different values of **H** were determined. In particular, it was shown that a ferromagnetic structure is realized in fields $H > H_F$, whereas the ferrimagnetic phase (3) is stable for $H_{FR}^{(1)} \leqslant H \leqslant H_{FR}^{(2)}$. For $H \leqslant H_{FR}^{(2)}$ a noncollinear angular phase arises, which goes over continuously into the ferromagnetic phase at $H = H_F$. We can identify the field H_F with the field H_{c3} observed in the experiment. It is more difficult to investigate the character of the phase transition for $H \leqslant H_{FR}^{(1)}$. Our calculations show that for $H_{c1} \leqslant H \leqslant H_{FR}^{(1)}$ the above-considered structures with spiral ordering of the components S_n in the plane perpendicular to the field **H** are stable in the range of values $k_F^{(3)} = 1.40$ Å$^{-1} \leqslant k_F \leqslant k_F^{(4)} = 1.45$ Å^{-1} (see Fig. 6). In this case, for all the spins, the projections on the direction of **H** are positive, and the magnitude of the spins increases with increase of the field. It is obvious that such structures cannot go over continuously into the ferrimagnetic phase (3). Therefore, we can assume that the appearance of the latter is a first-order phase transition, occurring in the field for which the energies of the spiral (with susceptibility $\chi_\perp$) and ferrimagnetic (3) structures coincide. In this case $H_{FR}^{(1)}$ determines the lower boundary of the intermediate state in which the spiral and ferrimagnetic phases coexist.

Thus, our analysis allows us to postulate that an external magnetic field induces in the compound $GdCu_2$ a whole cascade of spin-orientation transitions: First, for $H \geqslant H_{c1}$ ($\mathbf{H} \| \mathbf{a}, \mathbf{c}$), a rotation of the spins into the plane perpendicular to **H** occurs; upon further increase of the field ($\mathbf{H} \| \mathbf{a}, \mathbf{b}, \mathbf{c}$) a ferrimagnetic phase appears (by way of a first-order phase transition) and is stable up to the field $H_{FR}^{(2)}$; finally, for $H > H_{FR}^{(2)}$ a transition occurs from the ferrimagnetic phase (3) to the ferromagnetic phase and is completed in the field $H_{c3} = H_F$. As follows from the data of Fig. 7, this pattern of transitions can be observed in a rather narrow range of values of k_F ($k_F^{(1)} = 1.442$ Å$^{-1} \leqslant k_F \leqslant k_F^{(4)} = 1.45$ Å^{-1}); here the relative magnitudes of the fields $H_F = H_{c3}$, $H_{FR}^{(2)}$, and $H_{FR}^{(1)}$ (Fig. 8) are in reasonable agreement with the data that

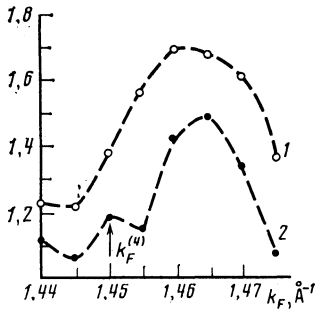

FIG. 8. Dependence of the relative magnitudes of the fields $H_F = H_{c3}$, $H_{FR}^{(2)}$, and $H_{FR}^{(1)}$ on k_F: $H_F/H_{FR}^{(1)}$; 2) $H_{FR}^{(2)}/H_{FR}^{(1)}$.

can be obtained from the magnetization curves (Fig. 4). The estimates of k_F performed in Ref. 3 in a rigid-band model give for $GdCu_2$ the value $k_F = 1.395$ Å^{-1}. With allowance for the approximate character of this estimate the range of values $k_F = 1.442$–1.445 Å^{-1} is highly probable for the $GdCu_2$ samples investigated.

CONCLUSION

Our investigations of the magnetic properties of the compound $GdCu_2$ show that the presence of a large magnetic-anisotropy energy is not a necessary condition for the realization of metamagnetic transitions: Such transitions are possible in the exchange approximation in those cases when there are ferrimagnetic structures whose exchange energy lies between the energies of the initial antiferromagnetic phase and the final ferromagnetic phase.

The transitions observed in $GdCu_2$ can be explained, at least qualitatively, in the RKKY-exchange model. The fact that the energies of the magnetic structures considered undergo considerable changes in response to relatively small variations of k_F makes it possible to conjecture that the magnetic properties of these compounds, and, in particular, the anomalies of their behavior in a field, will be extremely sensitive to the changes of conduction-electron density that occur, e.g., when Cu ions are substituted by Ni and Co ions.

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