Dynamic contribution to the magnetic anisotropy

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Anomalies in the angular and temperature dependences of the resonant field have been observed in the ferromagnet $HgCr_2Se_4$:Au. These anomalies can be explained in terms of the dynamic contribution to the cubic magnetic anisotropy. The reconstructed "static" dependences (those without this contribution) are devoid of such anomalies.

In this letter we report the results of a study of a ferromagnetic resonance in $HgCr_2Se_4$ crystals doped with 2 at.% gold and grown by the chemical transport-reaction method.[1] The polished spheres ~0.7 mm in diameter were oriented by an x-ray method within 1°. Figures 1 and 2 are plots of the angular and temperature dependencies, respectively, of the resonant field H_{res} and of the line width of the ferromagnetic resonance, $2\Delta H$.

The ferromagnetism of $HgCr_2Se_4$ arises due to the ordering of the magnetic moments of Cr^{3+} ions, while the magnetic anisotropy and relaxation are determined primarily by Cr^{2+} and Cr^{4+} ions which are produced because of the formation of vacancies and doping during the growth of the crystals. The feature that distinguishes $HgCr_2Se_4$ (Refs. 2 and 3) from $CdCr_2Se_4$ (Ref. 4), for example, is the coexistence of Cr^{2+} and Cr^{4+} ions in comparable quantities. It should be noted that actually complex centers which include Cr^{2+} and Cr^{4+} ions are formed and that at least at low temperatures the magnetic properties are determined by these ions. In Fig. 1 the H_{res} and $2\Delta H$ peaks in the ⟨111⟩ direction are caused[2,3] by the Cr^{2+} ions and in the ⟨100⟩ and ⟨110⟩ directions they are caused by the Cr^{4+} ions.

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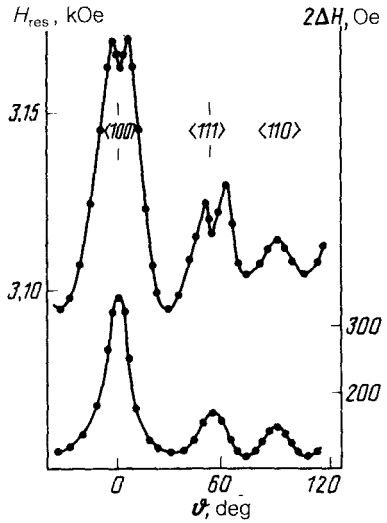

FIG. 1. Angular dependences of H_{res} and $2\Delta H$ in $HgCr_2Se_4$: 2 at.% Au, $T = 4.2$ K, $f = 8.9$ GHz, θ is the angle in the {110} plane between the constant field and the ⟨100⟩ axis.

The curves in Figs. 1 and 2 have the following systematic features which heretofore, to the best of our knowledge, have not been observed: the dips on the angular dependences H_{res} in the ⟨100⟩ and ⟨111⟩ directions and the low-temperature minima H_{res} for these directions. We will show below that these features can be explained by the dynamic (or relaxation) contribution to the crystallographic magnetic anisotropy.

The effect of rapidly relaxing ions, which the Cr^{2+} and Cr^{4+} ions are acknowl-

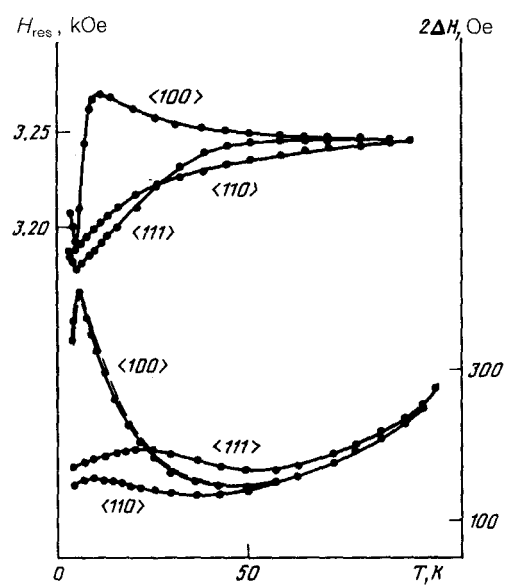

FIG. 2. Temperature dependences of H_{res} and $2\Delta H$ in $HgCr_2Se_4$: 2 at.% Au, $f = 9.1$ GHz. Dashed curve—calculation carried out for $\Delta\epsilon = 7.7$ K and $\tau_{i0} = 5\times10^{-11}$ s.

edged to be, with a strong spin-orbit coupling, on the magnetic anisotropy and magnetic relaxation in the case of a ferromagnetic resonance is generally described (see, e.g., Refs. 2–5) by means of the theory of "slow" relaxation[6] which is based on the concept of a delayed change in the populations of the energy levels of ions which are modulated by magnetization precession. According to this theory, the dynamic contribution of these ions in a resonant field is given by

$$\delta H_{\text{res}} = -\omega\tau_i \ \Delta H_i , \tag{1}$$

where ω is the angular frequency, τ_i is the relaxation time of the level populations, and ΔH_i is the ion contribution to the resonance curve half-width. To single out this contribution from the total line width, we assumed that it is the principal contribution at low temperatures and that it is negligible at the angular minima of ΔH.

Let us first consider the ⟨100⟩ direction. At low temperatures the temperature dependence τ_i can be approximated[7] in the following way:

$$\tau_i = \tau_{i0} \tanh \frac{\Delta\epsilon}{2kT} , \tag{2}$$

where $\Delta\epsilon$ is the spacing between two lower levels of the ion. The values of the parameters $\Delta\epsilon$ and τ_{i0} were determined from the temperature dependence of $2\Delta H$. At these parameter values this dependence was found to be described very well by the theory of slow relaxation (see Fig. 2). Using the functional dependence $\tau_i(T)$ found in this manner and the experimental values of $2\Delta H$, we calculated δH_{res} from Eq. (1) and we reconstructed the "static" values (after subtracting δH_{res}) of the resonant field $H^{\text{st}}_{\text{res}}$. The $H^{\text{st}}_{\text{res}}(T)$ curve (Fig. 3), which is a monotonic curve, is described well by the theory[6] without a dynamic contribution, i.e, a single-ion theory of anisotropy.

Working in a similar way, we reconstructed the angular dependence of $H^{\text{st}}_{\text{res}}$ under the assumption that τ_i changes only slightly in the case of a slight deviation of

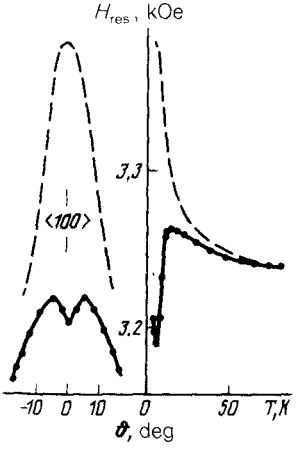

FIG. 3. A fragment of the angular dependence (near the ⟨100⟩ axis at 4.2 K) and the temperature dependence (for the same axis) of H_{res}. Points—experimental; dashed lines—reconstructed "static" values.

the field from the ⟨100⟩ axis (Fig. 3). This angular dependence consists of a single maximum, in agreement with the result of Ref. 6.

The contributions from the Cr^{2+} and Cr^{4+} ions are comparable in the ⟨111⟩ direction. To reconstruct H_{res}^{st}, a way must be found to separate these contributions, to determine the dynamic contribution for each one, and to sum over them. Such a calculation leads, as it does for the ⟨100⟩ direction, to a monotonic curve for $H_{res}^{st}(T)$ and to a peaking of $H_{res}^{st}(\theta)$ without a dip.

The "anomalies" of the angular and temperature dependences of H_{res} which we have observed can thus be explained by the dynamic contribution to the anisotropy according to the theory of slow relaxation. The other mechanism through which the ions with a strong spin-orbit coupling show their influence—the "fast relaxation"[8]—also leads to relation (1), but the calculated dynamic contributions in this case are too small. On the the other hand, the theory of slow relaxation, which describes well, as we have seen, the temperature dependence of $2\Delta H$ and the angular and temperature dependences of H_{res}, predicts the presence of sharp angular minima in the ⟨111⟩, ⟨100⟩, and ⟨110⟩ directions. As can be seen in Fig. 1, we have not detected these minima (these minima have also not been observed in many other studies). The mechanism for ferromagnetic relaxation in $HgCr_2Se_4$ and in similar crystals must therefore be refined, with allowance possibly for the complex structure of the centers or the electronic transitions between ions. Relation (1) would apparently remain valid, however, even after such a refinement.

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