MAGNETO-OPTICAL PROPERTIES OF GARNET FERRITES IN THE INFRARED REGION

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The Faraday effect at infrared frequencies was measured in yttrium and holmium garnet ferrites at room temperature and liquid nitrogen temperature. The sign of the Faraday effect in holmium garnet is reversed on passing through the compensation point, due to the rotation of the ferrimagnetic spin system in a magnetic field. An absorption band at $\lambda = 1.96\mu$ due to $^5I_8 \rightarrow {}^5I_7$ electronic transitions between ground-state sublevels of holmium ions was detected by both optical and magneto-optical means. By comparing the experimental data for yttrium and holmium garnets it was possible to detect the rotation of the plane of light polarization due to one of the magnetic sublattices of the ferrimagnetic substance.

GARNET ferrites, which are transparent in the infrared region, contain three magnetic sublattices (d, a, and c) consisting of 24 Fe^{3+} ions in d tetrahedral sites, 16 Fe^{3+} ions in a octahedral sites, and 24 rare-earth or yttrium ions in c sites.[1] The magnetic moment of the c sublattice is antiparallel to the total magnetic moment of the iron ions. Most garnet ferrites exhibit a so-called compensation point, the temperature at which the spontaneous magnetization of the garnet vanishes due to equality of the magnetic moments of sublattices c and d + a. At higher temperatures the magnetization of the iron sublattices is dominant, while at lower temperatures the magnetization of the rare earth sublattice predominates. A compensation point does not exist when the 24 c sites are occupied by nonmagnetic ions or by rare-earth ions with a small magnetic moment.

We investigated two garnet ferrites — $Y_3Fe_5O_{12}$ without a compensation point, the Faraday effect in which has been measured over a broad infrared region in earlier work,[2] and $Ho_3Fe_5O_{12}$ with a compensation point. Holmium garnet is very suitable for study because its compensation point is 136°K, so that approximately the same considerable amount of saturation magnetization is exhibited at room temperature and at liquid nitrogen temperature.

Figure 1 shows the distribution of the magnetic moments in the three sublattices of holmium garnet according to the Néel theory, and the orientations of these moments in an external magnetic field both above and below the compensation point. More detailed information concerning the crystallographic and magnetic structures of garnet ferrites can be found in Belov's monograph.[3] Since the yttrium ion is nonmagnetic, the magnetic moments of the d and a sublattices in Fig. 1 determine the absolute values of saturation magnetization in yttrium garnet. In other words, when we investigate yttrium garnet we are studying two magnetic sublattices that are contained in all garnets. In the other garnets the additional sublattice consists of magnetically active rare-earth ions.

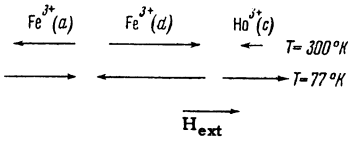

FIG. 1. Orientation of the magnetic moments of the three sublattices of holmium garnet ferrite in an external magnetic field. The lengths of the vectors are proportional to the magnetic moments of the sublattices.

Electrons in the well-screened f shell of rare-earth ions interact only weakly with the electrons of neighboring atoms. This circumstance facilitates the study of interactions in garnet ferrite crystals, since the energy level scheme of an isolated rare-earth ion can be used in zero approximation. Moreover, the weakness of the given reaction results in narrow absorption lines associated with transitions between f levels. The detection and study of narrow lines in solids, especially in ferromagnets, is important for quantum radiophysics.

The garnet samples were polished single-crystal plates about 100μ thick with about 0.1 cm^2 lateral face area. The monochromator was an IKS-11 spectrograph. Infrared light polarized by a selenium mirror passed through the single-crystal sample that had been magnetized perpen-

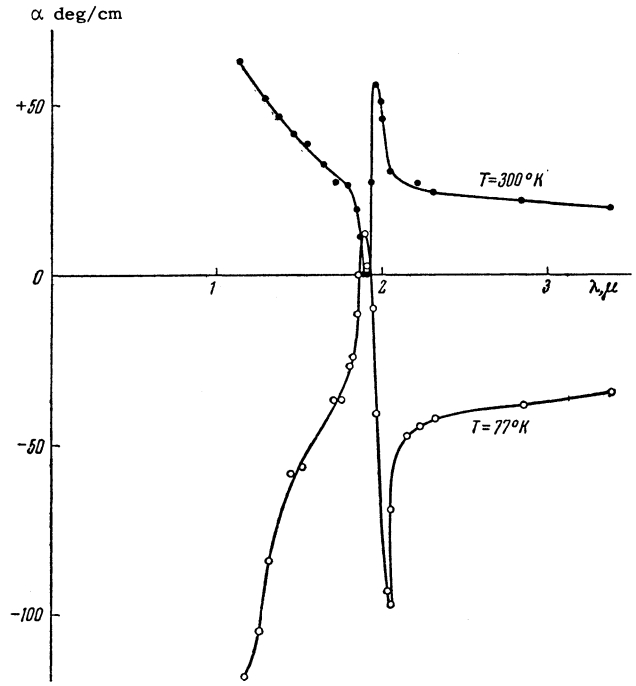

FIG. 2. Faraday effect in holmium garnet at two temperatures, above and below the compensation point. Ordinates represent the specific rotation α_Φ of the plane of polarization in degrees per cm.

dicular to its surfaces by a small electromagnet generating a field H = 1500 oe. The analyzer consisted of a few silver chloride plates[4] and was oriented at 45° to the polarizer. This angle yields maximum variation of transmitted light intensity as the plane of polarization is rotated.

For measurements at liquid nitrogen temperature the sample was placed in a cryostat with glass windows for light transmission. The light sources were a Nernst glower and a tungsten incandescent lamp; the detectors were a vacuum thermocouple and FÉU-22 photomultiplier. Absorption was determined by comparing measurements with and without the garnet. No correction was made for

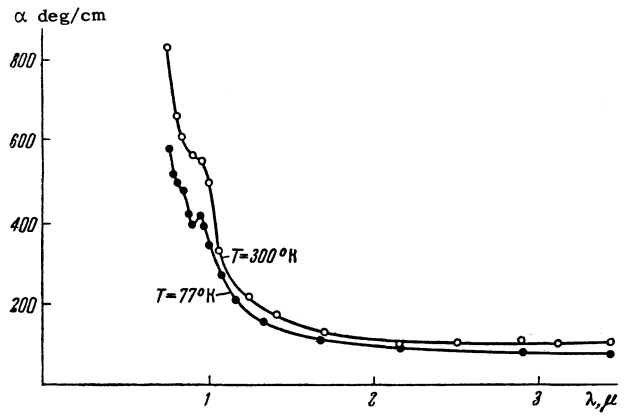

FIG. 3. Faraday effect in yttrium garnet at room temperature and at liquid nitrogen temperature.

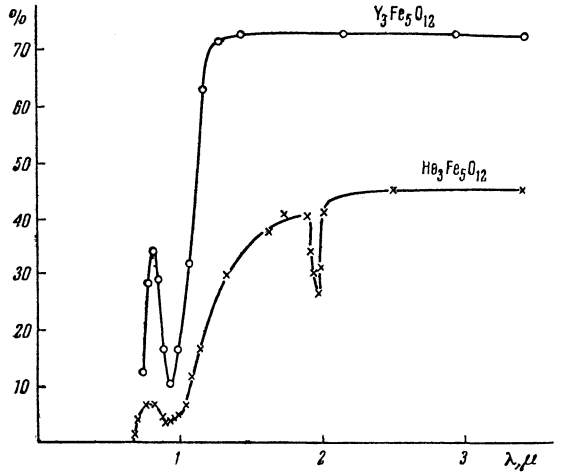

FIG. 4. Infrared transmission coefficients (%) of yttrium and holmium garnets.

reflection because of lack of data regarding the refractive index of garnets.

Figure 2 shows measurements of the Faraday effect in holmium garnet at two temperatures, above and below the compensation point. The curves are distinguished by the different directions in which the plane of light polarization is rotated by the garnet at room temperature and at 77°K. The reversal of the Faraday effect results from the reorientation of the atomic magnetic moments of all garnet sublattices on passing through the compensation point. Below this point the magnetic moment of the holmium sublattice, which becomes larger than that of the iron sublattices, is oriented parallel to the external magnetic field, and rotates the ferrimagnetic spin system through 180° (Fig. 1).

The observed effect is direct evidence for the correctness of the magnetic structure scheme of garnet ferrites proposed by Néel.[1] In addition, the sign reversal of the Faraday effect indicates that the rotation of the plane of light polarization is not associated with the resultant spontaneous magnetization of a garnet ferrite as a whole, which always assumes the direction of the external magnetic field, but rather with the magnetic moments of the separate ferrite sublattices. For example, measurements of the rotation of the polarization plane of microwaves revealed no reversal of rotation on passing through the compensation point, since this effect, like the great majority of physical effects in ferrimagnetics, depends on the resultant magnetic moment.

Figure 3 shows the Faraday effect in yttrium garnet at two given temperatures. The rotation of the polarization plane is not reversed because the 24 c sites are occupied by nonmagnetic ions.

Clogston[5] has attempted to formulate a theory of the Faraday effect in yttrium garnet. The characteristic frequencies of the dispersion curves of the Faraday effect are determined by forbidden transitions in Fe^{3+} ions. The oscillator strengths of these transitions and the magnitude of the Faraday effect depend on the existence of higher-lying levels to which electric dipole transitions are allowed. Therefore a quantitative check of the theory requires knowledge of the energy spectrum of iron ions in the garnet crystal.

The curves in Fig. 2 show a pronounced anomaly in the Faraday effect at $\lambda = 2\mu$. The measurement of absorption by holmium garnet showed that this anomaly is associated with the absorption band at $\lambda = 1.96\mu$ (Fig. 4). This absorption band is very probably attributable to an electronic transition ($^5I_8 \rightarrow {}^5I_7$) between the first two sublevels of the lowest multiplet level, which had been observed previously in Ho^{3+} ions at $\lambda = 1.94\mu$.[6] Yttrium garnet exhibits an absorption band at 0.92μ (Fig. 4), associated with an electronic transition in Fe^{3+}. Absorption by holmium garnet at wavelengths from 0.9 to 1.2μ is due to the indicated transition in Fe^{3+} and to the $^5I_8 \rightarrow {}^5I_6$ transition in Ho^{3+} ($\lambda_0 = 1.14\mu$).[6] Thus in the characteristic frequency region optical methods enable us to determine the fraction of light absorption and of the rotation of the polarization plane that is due to the ions of one of the garnet magnetic sublattices. This opens wide possibilities in the study of ferrimagnetics.

The 0.5-mm slits used for our measurements correspond to a wavelength interval of $\sim 0.05\mu$ at $\lambda = 2\mu$. It will be necessary in the future to improve the monochromaticity of light used in measuring the Faraday effect. Higher resolving power will enable us to improve the shape of the Faraday effect dispersion curve, and to study the fine structure and polarization of the separate components.

Figure 2 shows the measurements of the Faraday effect in holmium garnet only up to 3.5μ. Measurements at room temperature were carried to 5μ; the effect falls off slowly and gradually from 3.5 to 5μ. In both holmium garnet and yttrium garnet[2] monotonic variation of the Faraday effect will evidently be observed in the long-wave portions of the dispersion curves at visible and ultraviolet characteristic frequencies.

If it is assumed that in the present case the Faraday effect consists of three terms due to three magnetic sublattices, by comparing the curves in Figs. 2 and 3 we can distinguish the rotation of the polarization plane caused by the rare-earth sublattice consisting of holmium ions. Since the Faraday effect in holmium garnet is smaller but of the same sign as for yttrium garnet, the rotation of the polarization plane due to the holmium sublattice is smaller and opposite to that caused by the magnetic iron sublattices.

We have detected a similar reduction of the infrared Faraday effect in erbium and gadolinium garnets. The Faraday effect in yttrium garnet falls off with decreasing temperature (Fig. 3); this is evidently due to narrowing of the absorption lines of iron ions at lower temperatures. In holmium garnet the Faraday effect is enhanced with decreasing temperature. This is associated either with the occurrence of electronic transitions at 1.96 and 1.14μ, or with a sharper temperature dependence of the Faraday effect for the holmium sublattice than for the iron sublattices.

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