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IRREVERSIBLE PHOTONDOUCED CHANGE
IN THE OPTICAL ABSORPTION
OF YIG(Si) ON IRRADIATION

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Résumé. — Nous avons observé qu'à basse température le coefficient d'absorption \(\alpha\) du YIG dopé au Si\(^{4+}\) et au Fe\(^{2+}\) est modifié considérablement et irréversiblement sous l'effet d'une irradiation en lumière blanche. Pour Y\(_3\)Fe\(_{4.98}\)Si\(_{0.02}\)O\(_{12}\) à 1.15 \(\mu\) le coefficient d'absorption après irradiation est de 5 cm\(^{-1}\) plus petit que celui mesuré après refroidissement dans l'obscurité, i. e., \(\alpha'\) = 5 cm\(^{-1}\). La différence \(\alpha - \alpha'\) décroît graduellement à mesure que le cristal est réchauffé et devient nulle à 170 °K. On interprète le phénomène en supposant que les propriétés du Fe\(^{2+}\) au voisinage de Si\(^{4+}\) sont différentes des propriétés du Fe\(^{2+}\) quand il est éloigné de Si\(^{4+}\).

Abstract. — We find that at low temperatures the optical absorption coefficient \(\alpha\) of (Si\(^{4+}\), Fe\(^{2+}\)) doped YIG is changed substantially in an irreversible fashion by irradiation with intense white light. For Y\(_3\)Fe\(_{4.98}\)Si\(_{0.02}\)O\(_{12}\) at 1.15 \(\mu\) the absorption coefficient after irradiation is 5 cm\(^{-1}\) smaller than that measured after cooling in the dark, i. e., \(\alpha'\) = 5 cm\(^{-1}\). The difference \(\alpha - \alpha'\) decreases smoothly as the crystal is warmed, falling to zero at 170 °K. The phenomenon is interpreted in terms of the differing properties of Fe\(^{2+}\) close to Si\(^{4+}\) and Fe\(^{2+}\) far from Si\(^{4+}\) ions.

I. Introduction. — Two distinct classes of photoinduced effects have been seen in magnetic materials:

I. Irradiation at low temperatures by linearly polarized light has been found to alter the symmetry of several magnetic crystals (e. g., Y\(_3\)Fe\(_{4.96}\)Si\(_{0.04}\)O\(_{12}\)). The symmetry change was reversible, and has been seen in such fundamental properties as anisotropy, lattice constant, and optical dichroism [1, 2, 3].

II. Irradiation at low temperatures by unpolarized light has been found to alter the initial permeability and switching properties of several magnetic materials (e. g., single crystal and polycrystalline Y\(_3\)Fe\(_{4.96}\)Si\(_{0.04}\)O\(_{12}\)) [4]. These effects are irreversible. The low field phenomena have been interpreted as reflecting the interaction of domain walls and «pinning centers». We report the observation of a photoinduced change in optical absorption coefficient in YIG lightly doped with silicon. The irreversibility and silicon concentration level indicate that this is an effect of Class II. This irreversible photoinduced absorption, interesting in itself, appears to be widely useful in studying the atomic scale phenomena underlying all the photoinduced effects.

II. Samples and experiments. — Our crystals were all grown from PbO-B\(_2\)O\(_3\) flux using ultrahigh purity materials. Since we found that polarization had no influence on the change in absorption, sections of arbitrary orientation were used in some cases.

Absorption coefficients were derived from intensity measurements. To measure transmitted intensity, light from a monochromator was chopped mechanically, passed through the specimen, and detected by a cooled PbS cell and a lock-in amplifier. All intensity measurements were made with the specimen in a saturating magnetic field perpendicular to the axis of the optical system.

Measurements over a wide temperature range were made with the sample in a vacuum jacketed tube through which helium gas streamed. Cold helium from the boiling liquid and warm helium from a cylinder could be mixed in any proportion to vary the temperature from 300 °C down to about 8 °K. The output of a transducer affixed to the specimen card was scaled electronically to give a voltage proportional to temperature over this whole range.

III. Photoinduced absorption. — The phenomenon we report is most clearly seen in figure 1. There we show a recorder trace of the absorption coefficient \(\alpha\) as in \(I = I_0 e^{-\alpha x}\) versus temperature of a YIG crystal lightly doped with silicon as the crystal is cooled, irradiated and heated. The photon energy of the measuring light is 9 700 cm\(^{-1}\), definitely below that of the crystal field absorption at about 11 000 cm\(^{-1}\). The figure shows that \(\alpha\) varies little on cooling from ~ 300° to ~ 15°. At this point the detector was masked, and the sample subjected to 60 seconds of irradiation by an intense white light (130 mW/cm\(^2\)). During this irradiation the sample temperature did not rise significantly. On uncovering the detector, \(\alpha\) was found to have decreased by more than 5 cm\(^{-1}\). Though a small part of the effect relaxes in minutes, the change in \(\alpha\) shown here is essentially irreversible. As long as the specimen is kept at 15 °K, \(\alpha\) for 1.15 \(\mu\) is 10 cm\(^{-1}\).

As soon as the sample is warmed from the point C the absorption coefficient begins to increase. After reaching 90 °K, \(T\) was again decreased to about 15°, the point D on the figure. Note that cooling to D and warming from D, the curve retraces itself and runs parallel to the initial segment from A to B. Warming from D to 90°, \(\alpha\) does not change as it did on warming from C to 90°. Warming above 90°, \(\alpha\) again increases. The same sort of thing is seen when the crystal is cooled to points E and F. Finally, heating to about 170° completely wipes out the decrement in absorption coefficient, and the \(a(t)\) curve joins the AB segment.
IV. Interpretation. — The extra electron per silicon ion required for charge neutrality in YIG(Si) is confidently thought to reside on an iron in an octahedral site. Thus tetrahedrally coordinated Si$^{4+}$ give rise to octahedrally coordinated Fe$^{2+}$. Obviously, in a very lightly doped crystal it is possible to distinguish between (Fe$^{2+}$)$_{\text{near}}$ which are close to Si$^{4+}$ ions and (Fe$^{2+}$)$_{\text{far}}$ ions which are remote from Si$^{4+}$ ions, though in fact there must be gradations in remoteness. The photoinduced effects of Class II have been interpreted in terms of changes in the occupation numbers of these near and far sites [4]. Considered as sites accessible to the excess electron, these differ in the details of their energy level splitting and in their overlap with neighboring sites. Among other properties we can distinguish differences in ground state energy and in barrier height or localization energy. Because of the coulomb attraction between Fe$^{2+}$ and Si$^{4+}$, the ground state of (Fe$^{2+}$)$_{\text{near}}$ lies below that of (Fe$^{2+}$)$_{\text{far}}$.

On cooling, the excess electrons preferentially occupy the nearby sites; (Fe$^{2+}$)$_{\text{near}}$ predominates. On irradiation with light, the electron is excited to a higher state or band at about 10 000 cm$^{-1}$ from which it can decay to other sites [2]. In the irradiated state (Fe$^{2+}$)$_{\text{near}}$ is more important. Class II photoinduced effects reflect the fact that (Fe$^{2+}$)$_{\text{near}}$ and (Fe$^{2+}$)$_{\text{far}}$ are distinctly different dopants and contribute distinguishably different physical properties to the crystal. For instance, their substantially different optical absorptions presumably account for the difference between the absorption coefficients in the cooled state B and the irradiated state C of figure 1.

Heating of the irradiated sample to any temperature from 20° to 170°K annihilates or "boils away" a fraction of the (Fe$^{2+}$)$_{\text{near}}$ which were not eliminated by a somewhat lower temperature. Having boiled away, the electrons find their way to near sites. It appears that the localization energies range from at least as low as 20° up to 170°K. That such a range of sites should occur is not surprising. The coulomb field falls off slowly. At a concentration of 0.1 silicon atoms/formula, Si occupies one tetrahedral site in 300. For a uniform distribution, there is an Si$^{4+}$ ion in a cube with 6 or 7 sites along the edge. Thus truly remote octahedral sites will be relatively unusual, sites within three sites of a Si$^{4+}$ ion will be common. Adjacent site pairs and various kinds of near pairs of Si$^{4+}$ ions will be scattered liberally throughout the crystal. In addition, the situation is complicated by the fact that the crystal inadvertently includes some Pb$^{2+}$ (typically 0.01 atoms/formula) as well as other impurities.

V. Conclusion. — We have found that the optical absorption coefficient of certain silicon doped YIG compositions may be irreversibly decreased by irradiation with white light at low temperature. This is a new manifestation of one class of photoinduced effects previously seen only in low field permeability and switching properties. Studies of the variation of the absorption decrement on irradiation, $\alpha - \alpha'$, with thermal history demonstrate that absorption centers with a wide range of localization energies are important in the phenomenon. The dependence of the photoinduced absorption on such parameters as impurity concentration and wavelength enable us to study the phenomenon underlying this class of photoinduced effects. Such experiments will be reported elsewhere.

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