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OPTICAL PHENOMENA

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Research effort on the optical properties of magnetic crystals has taken an upward turn, as judged from the large number of interesting papers published in 1967. On the spectroscopy of magnetic insulators, much attention has been given to the optical effects connected with magnetic ordering. The main features of the optical magnon sidebands, of two-magnon infrared absorption, and of light scattering by magnons and magnetoelastic modes, are all basically understood. The effects of both magnetic ordering and exchange interactions on the optical spectrum of magnetic ions have also been the subject of many investigations.

On the materials side, perovskite-type magnetic crystals and semiconducting ferromagnets have interested many people. In particular, transparent ferromagnets and ferrimagnets often yield large magnetic circular dichroism and Faraday rotation in the visible. They are therefore of great importance from the practical point of view. Interest on garnets is apparently still alive.

Work on optical properties of ferromagnetic metals during 1967 has been mainly on the band structures for these metals. In addition to the usual spectroscopic technique and the Kerr-effect measurements, differential reflectivity measurements are now being applied to metals with excellent results.

This Chapter deals mainly with optical properties in magnetic crystals which undergo magnetic order-disorder transitions at certain critical temperatures. Because of the limited scope, optical effects related to paramagnetic impurities in non-magnetic crystals are generally not included here.

A. Optical Effects in Magnetic Insulators

1. Optical Effects Associated with Magnetic Ordering

The absorption spectrum of the $^6A_{1g} \rightarrow {}^4T_{1g}$ transition in MnF_2 shows two sharp magnetic-dipole lines of σ polarization and three broad magnon-induced electric-dipole sidebands of $\pi 1$, $\sigma 1$, and $\sigma 2$, [Greene et al. (1965)]. Sell et al. interpret the spectrum in terms of exciton-magnon excitations. The magnetic-dipole lines are attributed to the excitation of the k = o excitons, and the electric-dipole sidebands to the combined excitation of exciton and magnon with opposite wave vectors at the Brillouin zone edge. The selection rules, the lineshpae, and the temperature and magnetic-field dependence of these sidebands are shown to agree with experimental observation. The exchange mechanism proposed by Tanabe et al. (1965) to explain the two-magnon far infrared absorption in MnF_2 is believed to be responsible for the observed sidebands, as also suggested by Tanabe and Gondaira.

The occurence of these magnon sidebands of optical transitions is fairly general in antiferromagnets. McClure et al. have identified several magnon sidebands associated with magnetic-dipole transitions in MnF_2, FeF_2, CoF_2, $FeCO_3$, and $MnCO_3$. Magnon sidebands in $KMnF_3$ and in $CsMnF_3$ have also been observed by Aoyogi and by Stevenson, respectively.

Eremenki et al. have identified the absorption bands in $RbMnF_3$ and $KMnF_3$ of the $^6A_{1g} \rightarrow {}^4E_g$ transition of Mn^{2+} as due to exciton-magnon excitations involving either one or two magnons at the Brillouin zone edge. The intensity

of the two-magnon band is of the same order of magnitude as the one-magnon band. The effect of a magnetic field up to 200 kOe on these bands is explained in terms of magnetostriction.

Richards reports the far infrared absorptions by two-magnon excitations in MnF_2 (at ~ 100 cm^{-1}), FeF_2 (at ~ 155 cm^{-1}) CoF_2 (at ~ 120 cm^{-1}), and $MnCO_3$ (at ~ 52 cm^{-1}). An exchange mechanism is used to interpret the results. Halley suggests that the two-magnon absorption could also arise from indirect coupling of the spins to the orbital field through the infrared-active optical phonons via the exchange magnetostrictive part of the magnetoelastic interaction. Allen has observed, in the antiferromagnetic UO_2 for $T < T_N = 30.8$ K, absorption at 17.5, 79, 99, 110.5, 184, and 216.0 cm^{-1}. The lines at 79 and 99 cm^{-1} are shown to be non-degenerate antiferromagnetic resonance modes. The 17.5 cm^{-1} line is due to absorption by zone-boundary magnons. The others are anomalous phonon absorption induced by the strong magnon-phonon interaction. Aring and Sievers have also detected in UO_2 at 3 K these lines associated with antiferromagnetic ordering at 17.6, 79, and 100 cm^{-1}.

The problem of light scattering in magnetic materials received a great deal of attention. Scattering of light by magnons is first suggested by Elliot and Loudon (1963). Shen and Bloembergen (1966) have treated the problem in quantitative detail, and extended the calculation to scattering by magnetoelastic modes and to stimulated scattering. Theoretical aspects of spontaneous light scattering by magnons has been reviewed by Shen. The intensity of the one-magnon Raman scattering is estimated from the Faraday rotatory power of the magnetic ions, and is shown to agree with experiments in the case of FeF_2. Moriya has developed the theory by using the spin-dependent electric polarizability, with particular reference to antiferromagnetic iron group fluorides. Symmetry properties of this polarizability and the temperature dependence of the scattering intensity are discussed. The theory of light

scattering by magnetoelastic waves in ferromagnetic and antiferromagnetic materials has also been studies by Akhiezer and Bolotin, for case both far away from and close to magnetoacoustic resonance. The relation between the Faraday effect and light scattering by magnons has further been investigated by L'Vor, taking into account the rotation of polarizations of the incident and scattered radiation. L'Vor and Starobinets suggest the use of driven coherent spin waves to enhance the light scattering intensity. Auld and Wilson use the Faraday rotation data to estimate the intensity of light scattering by coherent spin waves in YIG. Genkin et al. discuss the various nonlinear susceptibilities in ferromagnets.

Experimentally, two-magnon Raman scattering is reported by Fleury et al. in MnF_2. The results are interpreted by the excited-state exchange coupling between sublattices. Dixon and Matthews have observed diffraction of light at 1.15μ by elastic waves in YIG. The results are used to evaluate the photoelastic tensor. Dixon has also noticed large acoustic Faraday rotation and group-velocity dispersion in the region where magnon-phonon interaction is strong. Smith has observed the diffraction of light at 1.15μ by the magnetoelastic wave at 1.1 GHz in YIG. He has found that the diffraction angle can be varied by the external magnetic field so as to change the magnon-phonon admixture in the magnetoelastic mode. Wang and Thomas have noticed that the threshold for spin-wave instability in YIG decreases in the presence of a Nd laser beam. This is described as the evidence of Raman excitation of coherent spin waves by the laser beam.

Casselman and Spectors discuss the effect of interaction of optical phonons with magnons on ferromagnetic and antiferromagnetic resonances in ionic crystals. They show that at the band edge, the dispersion curves of

magnon and optical phonon may cross in relatively low fields.

2. Optical No-Magnon Excitations in Magnetic Crystals

Ferguson et al. have demonstrated that exchange-coupled transition metal ions can be simultaneously excited to an excited electronic level. The two regions of absorption near 40,000 and 45,000 cm^{-1} in $KZn_{1-x-y}Mn_xNi_yF_3$ and $KMn_xNi_{1-x}F_3$ are due to simultaneous excitations of $Mn^{2+}(^6A_{1g} \rightarrow {}^4A_{1g}, {}^4E_g{}^a)$ and $Ni^{2+}(^3A_{2g} \rightarrow {}^1E_g)$, and $Mn^{2+}(^6A_{1g} \rightarrow {}^4E_g{}^b)$ and $Ni^{2+}(^3A_{2g} \rightarrow {}^1E_g)$ respectively.

Moch and Balkanski have investigated the transitions $^3A_2 \rightarrow {}^1E_a$ and $^3A_2 \rightarrow {}^1T_{2a}$ in NiF_2 near 15,000 and 21,000 cm^{-1} respectively. They find that a fine structure disappears above the magnetic ordering temperature (73.2 K). Yen et al. have noticed appreciable shift and broadening of the sharp magnetic-dipole transitions with temperature in MnF_2 amd FeF_2 from 2 K to 40 K. The effects are believed to be the result of exciton-magnon interaction. The shift is connected with the changes of magnetization and crystal lattice parameters. The broadening is attributed to Raman scattering of magnons by the exciton. Popov has considered theoretically how the exciton-magnon interaction would affect the position, shape, and width of the absorption bands and their temperature dependence.

Van der Ziel has studied the four sharp ^2E lines at 13747.0, 13769.5, 13909.5, and 13931.4 cm^{-1} in $Cr_2)_3$. The results are interpreted using the exciton theory. In an external magnetic field, the 13747 cm^{-1} line splits with a g factor of 4.62, and a new line is induced at 3.75 cm^{-1} away from 13769.5 cm^{-1} in the zero-field limit. This new line is interpreted as the result of the Davydov splitting (1962).

Optical properties of various transition metal ion fluorides with the perovskite structure (XYF_3; X = alkali ion, Y = transition metal ion) have been investigated by many authors. Nesterova et al. have studied the infrared absorption spectra of antiferromagnets $NaCoF_3$, $KCoF_3$, and $RbCoF_3$ in the range of 750 - 2,000 cm^{-1} at 77 and 295°K. All compounds show an absorption band at 1,200 cm^{-1} due to crystal field splitting of the Co^{2+} ground state ^4F. For $T < T_N$ the half width in $KCoF_3$ increases by 40 cm^{-1}, probably as a result of magnetic ordering. The crystal field effect is also used by Pisarev and Prokorohova to interpret the spectrum of $RbCoF_3$ between 4,000 and 28,000 cm^{-1}, and by Mehra and Vankateswarlu to interpret the visible spectrum of $RbMnF_3$.

Pisarev et al. and Shafer et al. independently find that the transparent $RbNiF_3$ is ferrimagnetic. Pisarev et al. have studied the spectrum of $RbNiF_3$ from 0.24 to 2μ at 77 and 295°K. They conclude that the main structure is due to crystal field effect, but the fine structure of some lines could be due to spin-orbit coupling. Chen et al. find that $RbFeF_3$ is also transparent in the visible with good optical quality. It is antiferromagnetic between 102 and 87°K, and becomes ferrimagnetic below 87°K due to change in the crystal structure. Boky et al. have noticed that $Rb(Ni_{1-x}Co_x)F_3$ with x = 0 - 0.3 is also a transparent ferrimagnet. It changes from green to dark red as x increases from 0 to 0.3, and changes from a magnetic easy plane to an easy axis.

Optical spectra of magnetically ordered oxides of perovskite structure containing rare earth ions have been investigated by Hüfner et al. on $TbAlO_3$, and by Faulhaber et al. on $ErFeO_3$, $HoFeO_3$, $DyFeO_3$, and $DyAlO_3$. Shifts and splitting of spectral lines due to magnetic ordering are observed.

Research on optical properties of garnets is continuing. Kolb et al. find that YIG grown from high-temperature-high-pressure aqueous solvents has high near-infrared transmission because of relatively small amount of impurities. Grünberg et al. deduce the crystal field levels of ^6F and ^6H of Dy^{3+} from the infrared and visible absorption and emission spectra of DyAlG and DyGaG. Gehring et al. notice that the shifts and splitting of the spectral lines in Dy aluminum garnet persist even above T_N (= 2.5°K), presumably due to local short-range magnetic ordering. Grant has studied the reflectivity spectra of YIG and YGG between 2 - 7.5 ev. The spectrum in YIG is interpreted as crystal field transitions of tetrahedral Fe^{3+} and charge-transfer bands. The spectrum in YGG is believed to arise from critical point transitions between O(2p) and Fe(3d) bands.

3. Optical Excitations Associated with Impurities in Magnetic Crystals

The impurity spectrum in a magnetic crystal is often affected by magnetic ordering. This problem has been considered theoretically by Krivoglaz and Levenson. The possible broadening of the zero-magnon impurity line and the possible appearance of magnon sidebands due to electron-magnon interaction is discussed. Gehring et al. find that in Eu^{3+}(1%) doped DyAl garnet, some fine structure of the Eu^{3+} lines, presumably induced by the magnetic structure of the garnet matrix, persists at temperatures higher than 10 T_N. The existence of such fine structure is believed to be due to short range order among Dy^{3+} ions.

Murphy and Ohlmann have shown that in Cr^{3+}-doped $GdAlO_3$ the two absorption lines and four fluorescence lines between 13,700 and 13,850 cm^{-1} can be described as due to the splitting of the ^2E and $^4A_2$ states of

Cr^{3+} by an isotropic exchange interaction between Cr^{3+} and Gd^{3+}. They deduced the exchange constants $J_{CR(^4A_2)-Cr(^4A_2)} = 2.1\ cm^{-1}$ and $J_{Cr(^2E)-Gd(^8S)} = 3.2\ cm^{-1}$ by assuming $J_{Gd-Gd} \sim 0$. Blazey and Burns also measure the fluorescence of Cr^{3+} in $GdAlO_3$ as a function of temperature. However, they find $J_{Cr-Gd} = 2.3\ cm^{-1}$ and $J_{Gd-Gd} = -0.06\ cm^{-1}$ by neglecting J_{Cr-Cr}.

Exchange-coupled impurity pair spectra in many crystals have been investigated. From the effect of uniaxial stress on the luminescence spectrum, Kaplyanskii and Przhevuskii have identified Cr^{3+} pairs of monoclinic, triclinic, and trigonal symmetry in ruby. With an applied electric field of 200 Kv/cm parallel to the c-axis, Kaplyanskii et al. notice that a large number of these pair lines have a symmetric doublet splitting, but others show no noticeable change. No observable effect is seen for $E \perp$ c-axis. From the measurements on the time constants of build-up and decay of luminescence under excitation by visible radiation and by electric fields, Gumlich et al. suggest that the emission bands at 7450 Å and 6350 Å in Mn-doped ZnS are due to Mn-Mn pairs. Exchange interactions between Ce^{3+} and its tetrahedral iron neighbors in YIG:1% Ce^{3+} with various substituted iron concentrations have been studied spectroscopically by Wickersheim and Buchanan. New spectral lines in the substituted garnets due to reduced exchange splittings on Ce^{3+} induced by the replacement of tetrahedral iron neighbors are observed.

Wood and Remeika find that the transmission window from 1μ to 6μ in YIG can be modified by changing the valence of iron ions or by doping with rare earth ions. The absorption below 6μ due to lattice vibration remains unchanged.

4. Magneto-Optical Effects in Magnetic Insulators

The dispersion of the Faraday effect in $RbNiF_3$ has been measured by Pisarev et al. and by Shafer et al. The structure can be explained by the relation between rotation and circular dichroism. Pisarev et al. find near the transition $^3A_2 \rightarrow {}^1E^a$ of Ni and below T_N, a remarkably large magnetic circular dichroism in $RbNiF_3$, but not in $KNiF_3$. The magnetic linear dichroism has the same order of magnitude in both crystals. The difference arises because $RbNiF_3$ is of ferrimagnetic structure and $KNiF_3$ is of antiferromagnetic structure. Kharchenko and Eremenko have measured the rotation in the antiferromagnets MnF_2 and $RbMnF_3$ at some visible frequencies in a field up to 150 KOe, and at 20 - 150°K. The frequency-independent part of the rotation is identified as due to antiferromagnetic resonance. Chen et al. find that $RbFeF_3$ has a large rotation in the uv and the visible (~ 1,000°/cm at 5,000 Å) with a low saturation field (250G). Measurements on Cotton-Mouton effect in $RbFeF_3$ are also reported.

Measurements on five rare earth garnets $Y_3Fe_5O_{12}$, $Gd_{1.5}Y_{1.5}Fe_5O_{12}$, $Gd_3Fe_5O_{12}$, $Dy_3Fe_5O_{12}$, and $Tm_3Fe_5O_{12}$ by Johnson between 77° and 300°K have shown that the Faraday effect at 1 - 4μ is associated with both electronic absorption and exchange resonance. Chetkin and Shalygin have investigated the temperature dependence of the rotation in TbIG between 25° and 350° K at 6.5μ where the contribution due to exchange resonance is dominant. They find that the rotation changes sign at 110°K when the contributions from different sublattices cancel one another. Belyaeva et al. have noticed that at 4.2°K and in a field up to 25 KOe, the intensity of the components of the $^4I_{\frac{15}{2}} \rightarrow {}^4S_{\frac{3}{2}}$ transition

of Er^{3+} in ErIG depends on the direction of polarization with respect to the magnetization. From the results, they find the anisotropy constants $K_1 = 9 \times 10^6$ erg/cm^3 and $K_2 = 5 \times 10^7$ erg/cm^3 for the crystal. Eremenko et al. have estimated an exchange field of $(2.6 \pm 0.2) \times 10^5$ Oe acting on Ho^{3+} from the observed rotation due to Ho^{3+} in YIG:Ho^{3+}. DeSorbo has shown that the Verdet constant in TbAlG, calculated from the observed rotation by taking into account the demagnetizing field, is proportional to the magnetic susceptibility. Daybell et al. have found that the low-temperature rotation in Terbium Alumina Silicate glass fits an equation for rotation in paramagnetic ions with a ground Kramers doublet.

The Faraday rotation of Eu^{3+} at low temperatures has been calculated by Sander and Kittel. A contribution from the Van Vleck paramagnetism is here partially cancelled by the diamagnetic term. The rotation should be observable in the microwave and infrared region. Briss and Shrubsall have considered theoretically how the polarizations of the electromagnetic waves would change as the waves propagate in a magnetoelectric crystal.

Teale and Temple have noticed a new magneto-optical effect, the change of magnetocrystalline anisotropy by infrared irradiation. The anisotropy of the silicon-doped YIG ($Y_3Fe_{4.9}Si_{0.1}O_{12}$) is changed by more than 100 Oe at 20°K. The change is interpreted as due to redistribution of excess electrons between iron ions.

B. Optical Properties of Magnetic Semiconductors

Magnetic semiconductors have received much attention because of their many attractive properties. However, little is known about the

nature of the optically excited electronic states or bands in these crystals. Mullen and Lawson have measured the reflectivity of pressed EuS powder from 2 to 14 ev. Significant structure is observed above the fundamental gap. Cho has used the APW method to calculate the spin-polarized electronic bands for EuS, and has shown the material should be ferromagnetic as well as semiconducting. The energy gap is found to be 1.51 ev as compared with the experimental value 1.645 ev. The transition corresponds to an indirect transition from the ground 3p shell in the S^{2-} ion to the excited 5d shell in the Eu^{2+} ion. Methfessel et al. have noticed that in EuO and EuS, the onset of ferromagnetic order is accompanied by an unusually large red shift of the visible absorption peak. This is interpreted by an exciton-like model in which the excited electron of Eu^{2+} is shared among its twelve nearest neighbors.

Another type of semiconducting ferromagnets of interest is the chalcogenide spinels of ACr_2X_4 (A = Cd or Hg, X = S or Se) reported earlier by Harbeke and Pinch (1966) and by Lehmann and Harbeke. At room temperature, $CdCr_2Se_4$ (T_c = 129.5°K) has the absorption edge at 1.32 ev and $CdCr_2S_4$ (Tc = 84.5°K) at 1.57 ev. Above Tc, the absorption edge shifts to higher frequency for lower temperatures. Below Tc, the edge shifts to higher frequencies in $CdCr_2S_4$ and to lower frequencies in $CdCr_2Se_4$. The shift is more in an external magnetic field. These optical properties are believed to be associated with magnetic ordering, and the edge shift above Tc due to short-range order.

Hüffman and Wild have measured the optical spectrum of the semiconductor-type antiferromagnet α-MnS (T_N = 152°K) from 0.02 to 14 ev. A shift

with temperature of the $^6A_{1g} \rightarrow {}^4A_1$ transitions is attributed to magnetic ordering.

Optical transmission and reflection have been used by Zanmarchi to show that MnTe is a semiconductor above and below T_N (= 307°K), and that the p-carriers move in a broad band. Chrenko and Rodbell have noticed that the infrared spectrum of CrO_2 shows no band gap and is consistent with its metallic-like electrical properties instead of semiconductor-like.

C. Optical Properties of Magnetic Metals

1. Theoretical Discussions

Connolly has calculated the energy band of Ni using the APW method within the framework of unrestricted Hartree-Fock scheme, in which the exchange terms are approximated by a local potential. Krinchik and Canshine have used a model with the energy difference $E(L_2') - E(L_{32}) < 0$ at the symmetry point L to evaluate the imaginary parts of the diagonal and the off-diagonal dielectric constant tensor of Ni in the infrared region.

A modification of the usual sum rule has been proposed by Phillips to account for the resonant character of the d-states in transition and noble metals. With the help of this partial sum rule, the optical spectrum of Cu has been calculated by Mueller and Phillips using the random phase approximation. The off-diagonal part of the conductivity in ferromagnetic metals has been calculated by Vedyaev and Kondorskii using the ladder-diagramatic technique.

2. Experimental Contributions and Interpretation

The technique of differential optical spectroscopy for achieving higher resolution has been extended to the case of transition and noble

metals. Low energy interband transitions (0 - 2 ev) in Ni detected by ac thermally modulated reflectivity by Hanus et al. show peaks at 0.25 and 0.4 ev and a shoulder at 1.3 ev due to transitions around the L point in the Brillouin zone. The results are interpreted as lending support to a band model with $E(L_2') - E(L_{32}) < 0$ at the L point. Gerhardt et al. have used piezo-reflectance measurements (ac pressure modulation) to identify optical electronic transitions and to evaluate the three deformation potentials in Cu.

The optical densities of states of Co, Ni, and Pd have been determined from photoemission measurements by Yu and Spicer. There is a large peak at about 5 ev below the Fermi level in Co, Ni, and Fe, but not in Pd. It is concluded that the peak is probably related to the ferromagnetism of the 3d transition metals. Photoemission measurements from 4.5 to 11.4 ev have been made by Breen et al. to determine the optical density of states down to 6 ev below the Fermi level in ferromagnetic Ni-Al alloys. The results are described in terms of the rigid band model and scattering phenomena.

Schröder and Önegüt have studied the visible and uv spectrum of Cu-rich Cu-Ni alloys at room temperature, and found that the main absorption edge remains unchanged with alloying, but disappears with 20 and 25% Ni. The results indicate that the optical properties of the alloys cannot be described by a rigid band model. Noskov and Sasovskaya have concluded from the infrared spectrum in Cu-Ni alloys that the absorption at $\hbar\omega > 0.44$ ev does not involve bands which are split by the s-d exchange interaction. Müller has noticed the similarity of the spectra of Eu and Ba between 0.3 and 4 ev. This suggests an almost identical band structure for the two metals.

The polar and transverse Kerr effects and the real and imaginary parts of the off-diagonal dielectric tensor elements for Ni, Co, and Fe have been determined between 0.22 and 6.0 ev by Krinchik and Artemjev. Ultraviolet magneto-optical resonances in these metals are detected. It is shown that the magneto-optical method can be used to separate contributions of the up and down spins to the interband optical transitions.

Afanas'yeva and Noskov have shown from the Kerr effect measurements on Ni and Co in the infrared that the spin-orbit interaction in these metals cannot be described by an effective field, as proposed by Voloshinskiy (1964). Afanas'yeva et al. has also concluded from the Kerr measurements on Ni from 0.8 to 15μ at room temperature that optical transitions start at 0.12 - 0.16 ev and the exchange splitting of the L_3-band should be 0.4 ev.

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