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Magneto-optics of II–VI diluted magnetic semiconductors

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K. S. Krishnan's pioneering contributions to the magnetism of condensed matter included insightful studies on crystals with 3d transition metal ions as one of the constituents. We illustrate, with examples of striking magneto-optic phenomena, how the incorporation of such ions in the tetrahedrally coordinated II–VI semiconductors (the II–VI diluted magnetic semiconductors) has created a splendid scientific opportunity. These II–VI magnetic semiconducting alloys exhibit huge excitonic Zeeman splittings which in turn result in a giant Faraday rotation and an impressively large Voigt birefringence.

As a Reader in Physics (1929–33) at Dacca University and as the first Mahendralal Sircar Professor at the Indian Association for the Cultivation of Science, Calcutta (1933–42), K. S. Krishnan initiated, developed and enriched several important lines of research on the magnetism of condensed matter. As an illustrative example, his paper entitled 'Magnetic studies on rhodochrosite, $MnCO_3$', co-authored with his student S. Banerjee, is fascinating[1]. Rhodochrosite – also known as dialogite – is isomorphous with calcite ($CaCO_3$) which has D_{3d}^6 symmetry. Unlike calcite, which is a clear and transparent crystal throughout the visible, a well-formed single crystal of rhodochrosite with no apparent defects or inclusions, displays an enchanting pink color due to the Mn^{2+} ions. The Mn^{2+} ions are located above and below the planar CO_3^{--} ions which are normal to the optic axis. The $^6S_{5/2}$ ground state of a free Mn^{2+} ion, dictated by Hund's rules, has a Landé g-factor = 2 and a large magnetic moment of 5.92 Bohr magnetons[2]. Crystal field effects for Mn^{2+} when incorporated in a crystal are expected to be small. Krishnan and Banerjee acquired well-developed, large, transparent crystals of rhodochrosite

and measured their large volume magnetic susceptibility and its temperature variation, as well as the feeble magnetic anisotropy expected for D_{3d}^6. The ingenuity and care exercised in the experimentation; the full recognition that the chemical purity of the naturally occurring crystals must be critically assessed; the complete mastery of the physical issues underlying the microscopic interpretation of the magnetic behavior of crystals, such as crystal field effects and Mn^{2+}–Mn^{2+} antiferromagnetic coupling – all of these cannot but command our admiration, now sixty years since the paper first appeared in print. And it is written with a command of English as well as an evident enjoyment and pride in the presentation of their accomplishments.

It is indeed a privilege to contribute to this special issue of Current Science dedicated to the birth centenary of K. S. Krishnan. In the context of the Krishnan–Banerjee paper on $MnCO_3$, it appeared particularly appropriate to review the spectacular magneto-optic phenomena displayed by $Cd_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xSe$, two of the tetrahedrally coordinated Mn-based II–VI ternary alloys. They are members of an extraordinary subset of II–VI semiconductors called Diluted magnetic semiconductors (DMS) in which the group II cations have been randomly replaced by a 3d-transition metal ion[3]. These ternaries occur with the zincblende or the wurtzite structure over large composition ranges. Among the DMS, it is the Mn-based ternaries which have been most intensely investigated. The ease of incorporating Mn^{2+} over a large composition range, the success in the growth of large single crystals, and the simplicity of the electronic structure of Mn^{2+} making experimental results amenable to theoretical analyses are the factors which have contributed to the focus on the zincblende $Cd_{1-x}Mn_xTe$ ($0 \leq x \leq 0.77$) and the wurtzite $Cd_{1-x}Mn_xSe$ ($0 \leq x \leq 0.50$)[3].

In our paper we concentrate on the striking manner in which the huge excitonic Zeeman effect and the associated Faraday and Voigt effect manifest in $Cd_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xSe$, and how the $Mn^{2+}-Mn^{2+}$ anti-ferromagnetic interaction leads to a large T_{AF}, the anti-ferromagnetic temperature introduced 'phenomenologically' in characterizing these magneto-optic phenomena. They are illustrated with examples from the research programme of one of the authors and his collaborators[4-12].

Excitonic Zeeman effect

The band structure of a tetrahedrally coordinated, direct gap, zincblende DMS, i.e. with T_d symmetry, has a zone center Γ_6 conduction band minimum and a Γ_8 valence band maximum. An external magnetic field $H \parallel \hat{z}$ (referred to an orthogonal right-handed coordinate system x, y, z) splits the Γ_6 level into two levels, corresponding to $\pm\frac{1}{2}$ spins, with energies $E_g \pm 3A$ and the Γ_8 level into four levels given by $\pm B$ and $\pm 3B$ corresponding to $\mp\frac{1}{2}, \mp\frac{3}{2}$ spins, respectively. Here the zero of the energy scale is defined by the valence band maximum at the zone center; E_g is the energy gap at $k = 0$; $A = (1/6)(\alpha M/g_{Mn}\mu_B)$; $B = (1/6)(\beta M/g_{Mn}\mu_B)$; α and β are the 's–d' and the 'p–d' exchange integrals for the conduction and valence band, respectively; M, magnetization per unit volume; g_{Mn}, Landé g-factor of the Mn^{2+} spins corresponding to the $^6S_{5/2}$ ground state of Mn^{2+}, and μ_B, the Bohr magneton. To a good approximation, the magnetization for a dilute system of Mn^{2+}, i.e. for small x, is given by $M = -g_{Mn}\mu_B x N_0 \langle S_z^{Mn}\rangle$, where $\langle S_z^{Mn}\rangle = (-5/2)B_{5/2}(\eta)$; N_0, the number of unit cells per unit volume; $\langle S_{Zn}^{Mn}\rangle$, the thermal average of the Mn^{2+} spins along the direction of the magnetic field; $\eta = (g_{Mn}\mu_B H)/(k_B T)$; k_B, the Boltzmann constant; T, the absolute temperature; and $B_{5/2}$, the Brillouin function B_J for $J = 5/2$. We note here that the Brillouin function, or equivalently M, shows a characteristic saturation for large (H/T). Figure 1 (i) shows the four Zeeman components a, b, c, d, into which the excitonic transition at E_0 splits as observed in the Faraday geometry, i.e. for light propagating along H. They occur at $E_a(-\frac{3}{2} \rightarrow -\frac{1}{2}) = E_0 + 3B - 3A$; $E_b(-\frac{1}{2} \rightarrow \frac{1}{2}) = E_0 + B + 3A$; $E_c(\frac{1}{2} \rightarrow -\frac{1}{2}) = E_0 - B - 3A$; and $E_d(\frac{3}{2} \rightarrow \frac{1}{2}) = E_0 - 3B + 3A$; the electric dipole allowed a and b transitions occur in the $\hat{\sigma}_+ = (1/\sqrt{2})(\hat{x} + i\hat{y})$ polarization whereas c and d occur in $\hat{\sigma}_- = (1/\sqrt{2})(\hat{x} - i\hat{y})$. Note, E_0 differs from E_g by the excitonic binding energy. In the Voigt geometry, i.e. for light propagation normal to $\hat{H}$, transitions a, b, c and d are polarized normal to $\hat{H}$, whereas two additional Zeeman components, e at $E_e(-\frac{1}{2} \rightarrow -\frac{1}{2}) = E_0 + B - 3A$ and f at $E_f(\frac{1}{2} \rightarrow \frac{1}{2}) = E_0 - B + 3A$, polarized along $\hat{H}$, are expected.

Figure 1 (i) also shows the crystal field splitting in

the valence band maximum of the zincblende symmetry, $\Gamma_8(T_d)$, into $\Gamma_9(C_{6v})$ and $\Gamma_7(C_{6v})$, the irreducible representations of the wurtzite symmetry (C_{6v}), whereas the $\Gamma_6(T_d)$ conduction band minimum of T_d corresponds to Γ_7 of C_{6v}. The Zeeman splittings of Γ_7, Γ_9 and Γ_7 for $H \parallel \hat{c}$, the six-fold symmetry axis of C_{6v}, and the $\hat{\sigma}_+$ and $\hat{\sigma}_-$ transitions relevant for the Faraday geometry with $H \parallel \hat{c}$ are indicated on the diagram.

In Figure 2, the excitonic signature of $Cd_{0.99}Mn_{0.1}Te$ at $H = 0$ (i) and its Zeeman splitting observed in the Faraday geometry with $\hat{\sigma}_+$ and $\hat{\sigma}_-$ polarizations (ii) and in the Voigt geometry, with the electric vector $E \parallel H$ and $E \perp H$ (iii) are displayed. The spectra in Figure 2 (ii) and Figure 2 (iii), recorded with wavelength modulation[13,14] at $T = 1.8$ K and $H = 15$ kG, clearly show the Zeeman components having polarizations entirely consistent with those predicted in Figure 1 for the Faraday and the Voigt geometry, respectively. In Figure 3, the magnetic field dependence of the energies of the Zeeman components conforms to $B_{5/2}(\eta)$ remarkably well, with $N_0\alpha = 0.22$ eV and $N_0\beta = -0.88$ eV. The power of wavelength modulation enables the Zeeman components to be identified and their polarization characteristics to be delineated without ambiguity. The large values for $N_0\alpha$ and $N_0\beta$ which characterize DMSs should be underscored.

Faraday effect

As is well known, a linearly polarized light traversing an optically inactive isotropic medium placed in an
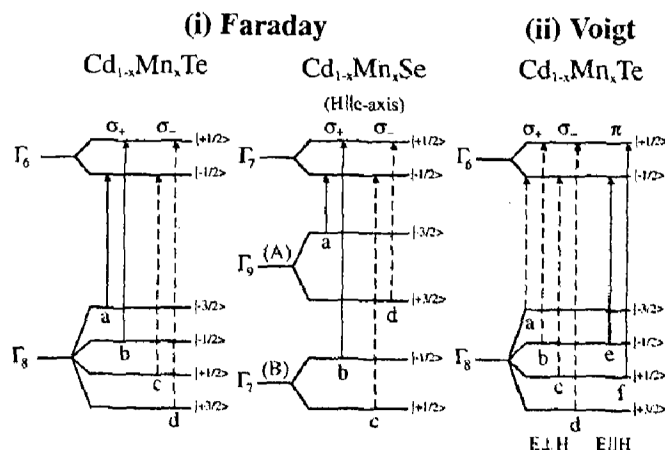
(i) Faraday

$Cd_{1-x}Mn_xTe$ $Cd_{1-x}Mn_xSe$

(ii) Voigt

$Cd_{1-x}Mn_xTe$

Figure 1. i) Schematic diagram of conduction and valence band splittings and interband transitions at the zone center of the zinc-blende $Cd_{1-x}Mn_xTe$ and the wurtzite $Cd_{1-x}Mn_xSe$ diluted magnetic semiconductors in the Faraday geometry. (ii) The transitions observed in the Voigt geometry for $Cd_{1-x}Mn_xTe$. Note the a, b, c and d transitions of $Cd_{1-x}Mn_xTe$ in the Faraday geometry are labeled 1, 3, 4 and 6, respectively; in the Voigt geometry, the transitions 2 and 5, corresponding to e and f (defined in the text), appear only in the Voigt geometry.

external magnetic field **H**, experiences a rotation in its plane of polarization. This magnetic-field-induced rotation Θ_F is called the Faraday rotation and the phenomenon is labeled the Faraday effect[15]. The angle Θ_F is proportional to the distance traversed in the material ℓ and the magnetic field **H**; the constant of proportionality V is the Verdet constant (the rotation per unit length per unit magnetic field). The phenomenological explanation of the Faraday effect closely follows that given by Fresnel for natural optical activity. A plane polarized wave transversing the medium, viewed as a superposition of $\hat{\sigma}_+$ and $\hat{\sigma}_-$ waves of opposite helicities, suffers circular double refraction. In other words, the two circularly polarized components propagate with different velocities, one faster and the other slower than the velocity in zero magnetic field. The rotation is given by the Fresnel formula $\Theta_F = (E \ell / 2\hbar c)(n_- - n_+)$, where $n_\mp$ are the indices of refraction for $\hat{\sigma}_\mp$ polarized light, E is the photon energy of the incident radiation, and c and $\hbar$ have the usual meaning. The microscopic origin of the circular birefringence, $n_- - n_+$, is the Zeeman effect causing the

difference in the dispersion of n_+ with respect to that of n_-, the former being associated with the $\hat{\sigma}_+$ polarized and the latter with the $\hat{\sigma}_-$ polarized Zeeman transitions.

In the case of $Cd_{1-x}Mn_xSe$, one has to contend with the linear birefringence intrinsic to the wurtzite structure. If the direction of light propagation and of **H** are restricted to be along the optic axis $\hat{c}$, the experimental observations[7] and their interpretation are then free from this complication and the Zeeman components relevant in the Faraday effect are the $\hat{\sigma}_+$ polarized a and b transitions and the $\hat{\sigma}_-$ polarized c and d shown in Figure 1 (i).
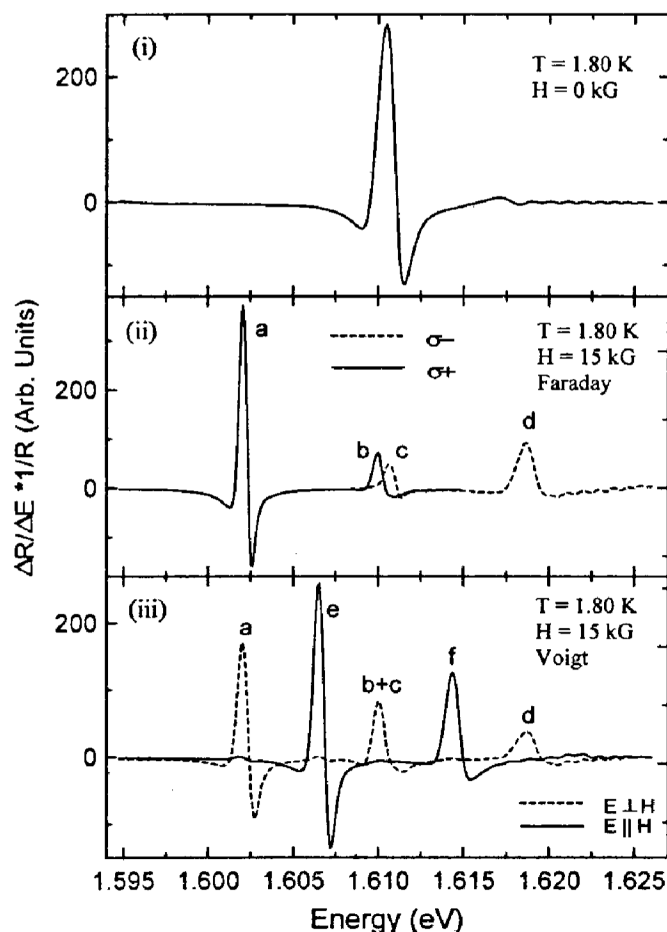

Figure 2. (i) Excitonic signature of $Cd_{0.99}Mn_{0.01}Te$ in the wavelength modulated reflectivity, recorded in the absence of a magnetic field. (ii) Zeeman effect of the excitonic signature in the Faraday geometry. (iii) Zeeman effect of the excitonic signature in the Voigt geometry.
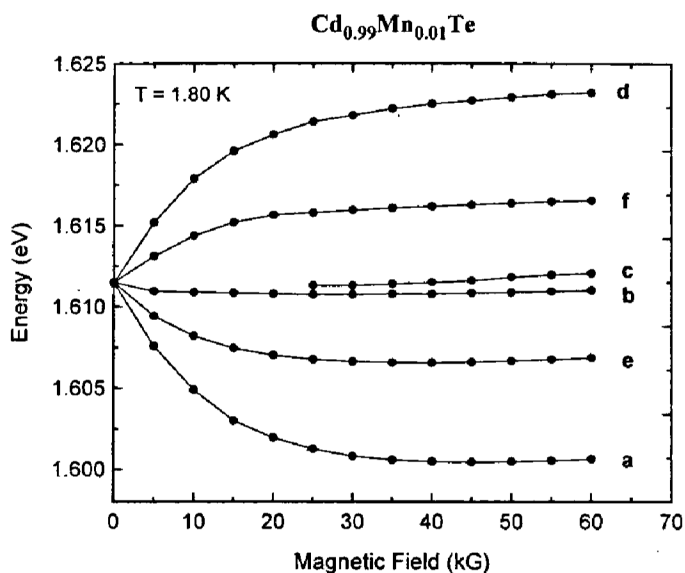

Figure 3. The energies of the excitonic Zeeman components of $Cd_{0.99}Mn_{0.01}Te$ at 1.8 K recorded as a function of applied magnetic field. The labels a, b, c, d, e and f correspond to those in Figure 1.

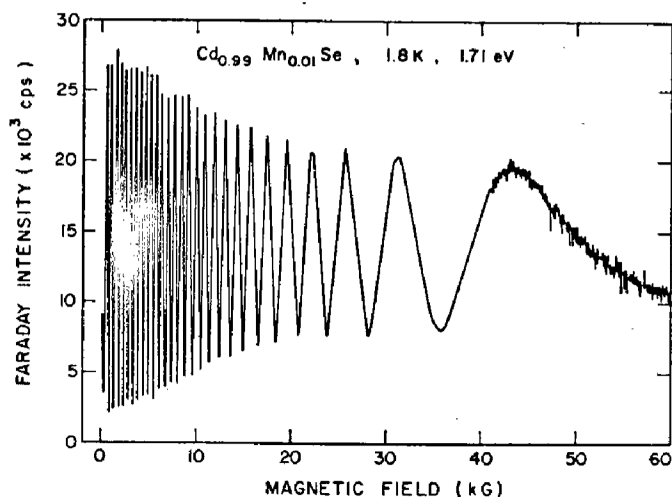Figure 4. 'Faraday' oscillations in the intensity of light at a fixed photon energy = 1.71 eV transmitted through $Cd_{1-x}Mn_xSe$ ($x = 0.01$) along its optic axis as a function of H. Thickness = 2.9 mm and $T = 1.8$ K (ref. 7).

In Figure 4 we display the oscillations in 'white' light spectrally analysed after it has traversed a slice of $Cd_{0.99}Mn_{0.01}Se$ along its optic axis, held between a polarizer and an analyser at $\pi/4$ with respect to each other[7]. Whenever $\Theta_F = m\pi + \pi/4$ for a given frequency, m being an integer, the transmitted intensity – the Faraday intensity – is a maximum; each successive maximum in the Faraday intensity observed with increasing H represents an additional π rotation. For low magnetic fields, the period of the oscillations is constant, whereas for high magnetic fields this period increases dramatically as the magnetization saturates. The Faraday rotation Θ_F as a function of H, shown in Figure 5, can be fitted to $B_{5/2}(H/(T + T_{AF}))$ with Θ_F (saturation) $= -5232°$ and $T_{AF} = 0.5$ K, thus showing that even for a very dilute DMS alloy the exchange coupling between Mn^{2+} ions is manifested. We also notice in Figure 5 that the amplitude of the Faraday oscillations decreases with

increasing H, demonstrating the ellipticity generated by the unequal absorption of $\hat{o}_+$ and $\hat{o}_-$, i.e. the *circular dichroism* associated with the closer and closer proximity of the photon energy to the $\hat{o}_+$ transitions, in contrast to the $\hat{o}_-$ transitions which move away from it.

Voigt effect

The Voigt effect[9] is yet another spectacular magneto-optic phenomenon displayed by DMS, with the huge excitonic Zeeman effect being the underlying microscopic mechanism. Consider a linearly polarized monochromatic light incident normally on an otherwise isotropic material (e.g. $Cd_{1-x}Mn_xTe$) placed in a magnetic field applied perpendicular to the wave vector of the radiation. For experimental simplicity let the electric vector E be polarized at $\pi/4$ to H. It can be shown that the transitions polarized $\perp$ H (a, b, c, d), i.e. $\hat{o}_\pm$ transitions, are associated

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The other side of the physicist

In his personal reminescence commemorating the 60th birth anniversary of KSK, C. Mahadevan, former Principal of Andhra University College, Waltair, writes, 'Krishnan was a great football fan. Our work in the laboratory kept us nearly late and in the case of Krishnan he would probably be engaged in talks with Prof. Raman. As soon as we could get out, we would shout for a taxi and rush to the Eden Gardens for the football match, paying black market prices for the tickets as we were always late. Krishnan was one of the most vociferous spectators, shouting, gesticulating, cheering and giving running commentaries during the progress of the play, much to the amusement and sometimes to the annoyance of the bystanders. His child-like simplicity manifested itself on such occasions.' KSK also loved playing tennis, badminton and bridge.

Here is an excerpt from the NIAS-Bangalore handout, distributed during the 100th anniversary celebration: 'For those of us who were with him during the period, we were all amazed by his broad knowledge of European literature, European art and particlarly by his sense of humour. He was at home talking to the famous Ballerina Ullanova, as much as he was talking to Kapitza. At one of the Soviet parties, a lot of vodka was being served with many toasts. Naturally, as a teetotaler, Krishnan would not touch it. The Russians were, however, telling the team that vodka should be drunk in one gulp and not sipped like the other kinds of alchoholic drinks. But with his usual sense of humour Krishnan said I will tell you an even better way to drink and that is 'the Madras way'; and he poured the vodka straight into his mouth without the glass touching the lips. The Russians were amazed and those, who were with him, were worried as he was a teetotaler and the drink could be bad for him. He, however, was as merry as could be and the evening was a great success.'

Philosophy, religion and literature were also dear to KSK's heart. In an article commemorating KSK's 60th birthday, K. R. Ramanathan wrote: 'Believing as Krishnan does that science alone cannot solve all problems in life, he combines an earnest study of the classics of Indian religious liternature with the study and practice of science, and endeavours to live a life of religious faith. The combination of *Vinaya* (humility) with *Vignyan* (critical understanding), which has become natural to him, makes it easy for people of all stations in life to feel at ease in his company.

'It is not surprising that Krishnan is much sought after for giving addresses and discourses not only on scientific but also on literary and philosophical subjects. He has great respect for Indian scholars of Sanskrit and other Indian languages who have been brought up in the traditional Indian way. Being himself a writer of distinction on philosophic and scientific subjects in Tamil, Tamil and Sanskrit scholars all over India and in Ceylon find him congenial and stimulating company, and take delight in inviting and honouring him at their conferences and ensembles.'

One of KSK's grandsons Thiruvadi Vijayaraghavan, who lives at Bangalore, has a collection of KSK's writings and correspondences that go beyond science. Vijayaraghavan informs me that they are in the process of bringing out a biography of KSK in this centenary year.

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with the dispersion of the refractive index for $E \perp H$, defined as $n_\perp = n_\sigma$; similarly the e and f transitions polarized along H influence the dispersion of $n_\parallel = n_\pi$, the refractive index for $E \parallel H$. Here e and f are labeled as the π transitions in Figure 1 (ii). It can be readily shown[9] that the phase difference between the two components of E resolved parallel and perpendicular to H, respectively, develop a phase difference ϕ given by $\phi = YH^2 l = E l /\hbar c)(n_\perp - n_\parallel)$ as they exit the sample. The parameter Y is the phase difference per unit length per unit magnetic field squared and E, the photon energy. Being second order in H, the Voigt birefringence is usually small in non-magnetic semiconductors and observable only for photon energies very close to the band gap. In contrast, in the DMSs it is enormous for
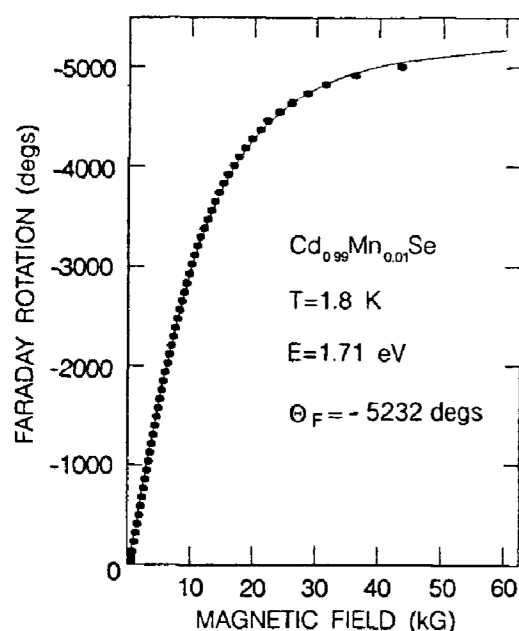
the same reason as is the Faraday effect, namely, the exceptionally large magnetization, M. However, it differs from the Faraday effect in its proportionality to M^2 rather than to M. Again, the $B_{5/2}(\eta)$ dependence of M enables one to enhance ϕ for high magnetic fields and low temperatures.

If the linearly polarized monochromatic radiation transmitted by $Cd_{1-x}Mn_xTe$ in the Voigt geometry is now analysed with an analyser at $-\pi/4$ with respect to H, it is easily seen that the transmitted intensity would exhibit maxima for $\phi = (2m+1)\pi$ and minima for $\phi = 2m\pi$, m being an integer. In Figure 6 the variation in the transmission of $Cd_{1-x}Mn_xTe$ ($x = 0.35$) is displayed as the magnetic field increases from 0 to 60 kG at $\hat{v} = 16,500 \, cm^{-1}$. The H^2 dependence of the Voigt effect, demonstrated in Figure 7 a, is obtained from the data for Figure 6. The magnetization of DMS follows the

Figure 5. Faraday rotation (Θ_F) versus magnetic field in $Cd_{1-x}Mn_xSe$, $x = 0.01$ at $T = 1.8$ K. The solid line is a Brillouin function, $B_{5/2}(H/(T + T_{AF}))$, fit to the magnetic field dependence of Θ_F, with Θ_F (saturation) $= -5232°$ and $T_{AF} = 0.5$ K (ref. 7).

Figure 7. The phase difference ϕ, (produced by the Voigt effect) as a function of H^2 for $Cd_{1-x}Mn_xTe$, (a) $x = 0.35$, at $T = 20$ K and $\bar{v} = 16,500 \, cm^{-1}$ and (b) $x = 0.18$, $T = 5$ K and $\bar{v} = 14,000 \, cm^{-1}$, demonstrating the M^2-dependence of the Voigt effect, where M is the magnetization. The saturation in ϕ is due to the saturation of M at high magnetic fields. As can be seen, for the lower x and T, this saturation effect is more pronounced (ref. 9).
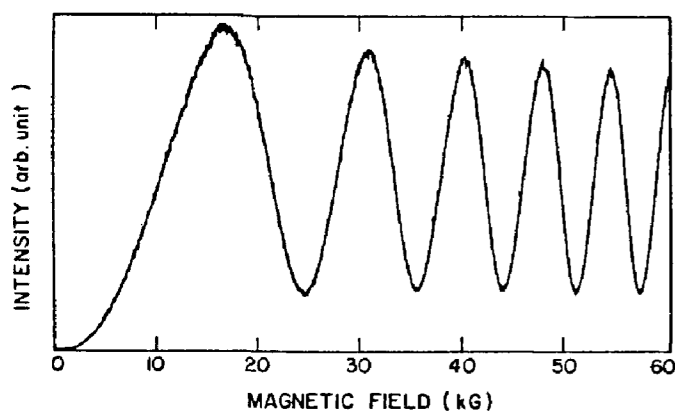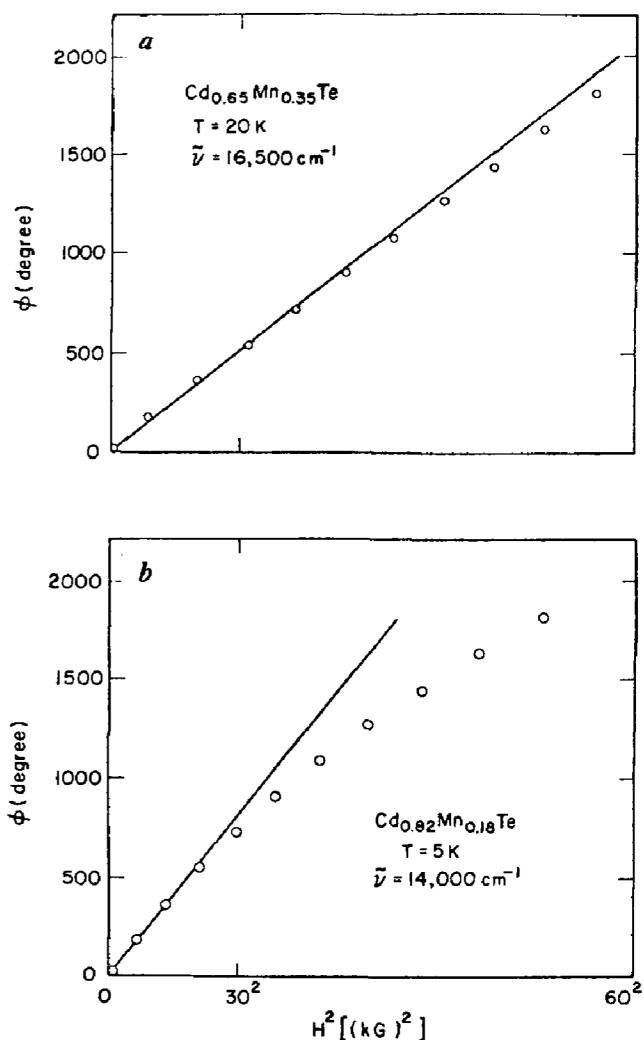
Figure 6. Transmitted intensity vs H, showing the magnetic-field dependence of the Voigt effect for $Cd_{1-x}Mn_xTe$, $x = 0.35$, $T = 20$ K, $\bar{v} = 16,500 \, cm^{-1}$ (ref. 9).

Brillouin function $B_{5/2}(H/(T + T_{AF}))$ in the low temperature regime; T_{AF}, the phenomenological measure of the $Mn^{2+}-Mn^{2+}$ antiferromagnetic interaction, is large for $x = 0.35$. The departure from linearity of ϕ vs H^2, expected from the $B_{5/2}$ function, thus manifests only at the higher fields; for another specimen with $x = 0.18$ and $T = 5$ K, this precursor to saturation at high fields sets in markedly at a lower value of H (Figure 7 b).

Although Voigt effect, a magneto-optic phenomenon second order in H, is usually very difficult to observe at laboratory magnetic fields for non-magnetic semiconductors, it is readily accessible in DMSs; indeed, for photon energies near the band gap and large H, one can match the natural birefringence of calcite in the visible range with the Voigt birefringence of a DMS.

Concluding remarks

Besides the three phenomena discussed in this paper, additional magneto-optic phenomena in the DMSs, traced to the large 'sp–d' exchange interaction and to the $Mn^{2+}-Mn^{2+}$ antiferromagnetic coupling (often referred to as the 'd–d' interaction), have been discovered. (1) *Raman–electron paramagnetic resonance (Raman–EPR)*[16]: The electronic transitions within the Zeeman multiplet of Mn^{2+} and Co^{2+} in $Cd_{1-x}Mn_xTe$, $Zn_{1-x}Mn_xTe$, $Cd_{1-x}Mn_xSe$, $Cd_{1-x}Mn_xS$, $Cd_{1-x}Co_xSe$, ... have been observed in inelastic light scattering as Raman shifts, hence the label Raman–EPR. They have also been observed in combination with the longitudinal optical zone center phonons of the DMS ternaries and quaternaries (both CdTe-like and MnTe-like LO-phonons in $Cd_{1-x}Mn_xTe$, for example). (2) *Raman–anti-ferromagnetic-resonance (Raman–AFMR)*[17]: The collective oscillation of Mn^{2+} spins in a DMS in its antiferromagnetic phase appears in the Raman spectrum as a 'magnon' feature and exhibits a Zeeman splitting; its relationship to the Raman–EPR in the paramagnetic phase has been studied and delineated[16]. (3) *Spin-flip of the donor-bound electron*[18–24]: The spin flip of donor-bound electrons in a DMS manifests as a Raman line with a large shift, which follows the characteristic $B_{5/2}(\eta)$ behavior associated with the large 's–d' interaction of the electron with its large orbits described in the effective mass theory of donors. The remarkable observation of the so-called bound-magnetic-polaron, *i.e.* a finite Raman-shift at zero magnetic field, is a unique feature extensively studied experimentally[19,22,24] and theoretically[25,26]. (4) *The Raman spectroscopy of multiple quantum wells and superlattices grown by molecular beam epitaxy*[27,28]: The magnetic excitations in the sub-micron heterostructures of DMSs grown by MBE has been extremely fruitful in the discovery of new aspects related to the reduced dimensionality.

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