Optics of absorbing anisotropic media

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IT is well-known that a plate of tourmaline cut parallel to the optic axis has a transparent green appearance while a plate of the same material cut perpendicular to the optic axis looks almost black. This is due to the fact that the ordinary ray is almost completely absorbed while the extraordinary ray is weakly absorbed. This property of tourmaline is called *dichroism*. As tourmaline belongs to the uniaxial class, only these two principal sections are important. But in a biaxial crystal the colour of the crystal can vary drastically as we go from one principal section to another. This phenomenon is called *pleochroism* or multicolouredness. The well-known pleochroic crystals are iolite, epidote, andalusite and titanite. Pancharatnam[1] started his research career with a study of the pleochroism in amethyst. Interestingly, the contributions for which Pancharatnam is well-known are the fall out of his investigations into the optics of absorbing crystals.

In this article we address ourselves to some important phenomena, encountered in an anisotropic medium, within and in the neighbourhood of its absorption bands. After highlighting the optical effects of absorption discovered by Pancharatnam we will pay attention to magneto-optics of absorbing and metallic crystals. In absorbing crystals in the neighbourhood of an absorption band we find many interesting effects due to spatial dispersion. A few of these effects will also be briefly discussed.

Absorbing biaxial crystals

Absorbing crystals, in a section perpendicular to the optic axis, have many interesting properties in conoscopy, i.e. in convergent or divergent illumination (see Box 1). For example, in transmitted natural light (i.e. without polariser or analyser) one has in uniaxial crystals either a bright central spot (corresponding to the optic axis) surrounded by a dark field or a dark central spot surrounded by a bright field depending upon whether k_o the absorption coefficient for the ordinary ray is less or more than k_e the absorption coefficient for the extraordinary ray. However, in biaxial crystals we get a dark brush – Brewster's Brush – through the point representing the optic axis and along a direction perpendicular to the axial plane. The brush is intercepted by a bright spot (corresponding to the optic axis) and exhibits a fringe system of low visibility. On the other hand, with polariser or analyser alone, in uniaxial crystals, one finds a dark brush intercepted by a central bright spot for $k_o < k_e$, the brush being parallel to the direction of incident vibration. For $k_o > k_c$ crystals, the dark brush has no bright spot and is perpendicular to the direction of incident vibration. In biaxial crystals we find either a continuous dark brush in the axial plane or a dark brush with a bright spot (corresponding to the optic axis) along a direction orthogonal to the axial plane, depending upon the relative magnitudes of the principal absorption coefficients. Also in biaxial crystals the brushes are intercepted by a bright and dark fringe system – idiophanic rings.

Dichroism and Poincaré sphere

Many of these features can be accounted for on the basis of the electromagnetic theory[2,3]. Absorbing crystals will have to be described by two types of tensors, viz. the index tensor and the absorption tensor. Both the tensor surfaces should confirm to the symmetry of the crystal. For example, they are ellipsoids of revolution about the optic axis in uniaxial crystals and triaxial ellipsoids with coincident principal axes in orthorhombic crystals. Pancharatnam[4] showed that in such crystals, locally, the effects of linear phase retardation δ, optical rotation ρ, linear dichroism k and circular dichroism σ are superposable and that the central sections of the index and absorption ellipsoids would be important in an analysis of light propagation. He solved the problem by employing the Poincaré sphere representation (see Box 2). We know[5] that to get the emergent state in the case of a linear (or circular) birefringent medium the polarization state P, representing the incident light, is rotated through δ (or ρ) about the line connecting the linear (or circular) orthogonal base states X_r and Y_r that propagate unaltered through the medium. For linear (or circular) dichroism alone, Pancharatnam[4] gave the following recipe:

Let X_k and Y_k represent, in the absence of linear (or circular) birefringence, the states of principal linear (or circular) orthogonal vibrations. These are absorbed to different extents. Then any incident state of polarization P gets resolved into these base states. The state of polarization on emergence, from a sample of thickness dz, is given by shifting the state P towards the less absorbed component, X_k say, along a meridian passing through X_k, P and Y_k by an amount $ds = (k \sin 2\xi)\, dz$ with 2ξ as the arc length $X_k P$.

Box 1. Conoscopy-interference phenomena in polarized light

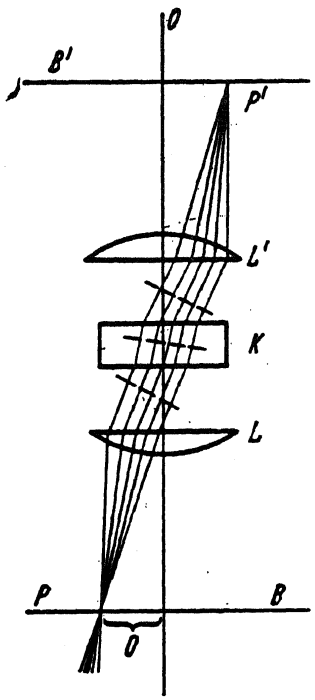
A method of observation of interference between polarized beams is by the use of 'convergent light', with the aid of a conoscopic arrangement shown in the figure. In this case the interference effects occur at infinity, i.e. at the focal plane of the lens. Each point in the focal plane is a focal point of a bundle of parallel rays emerging from the crystal in a particular direction. Since the retardation introduced by the plate varies with direction, the interference phenomenon varies over the field of view. The figure shows one particular bundle of parallel rays incident on the crystal. The incident ray gets resolved into two orthogonal linear vibrations on entering a transparent anisotropic optically inactive crystal. The path retardation suffered by each vibration should be calculated using the phase velocity. If n_1 and n_2 are the refractive indices and r the angle of refraction of the wave normal then the difference in the phase retardation suffered by the two components is to a good approximation given by:

$$\Delta = \frac{2\pi (n_1 - n_2)}{\lambda_0 \cos r} t,$$

where t is the crystal thickness and λ_0 the wavelength of light.

In absorbing systems the incident light gets resolved inside the medium, into permitted non-orthogonal base states which travel with different velocities and amplitude reductions. The state of the emergent light is obtained by adding coherently the base states with their appropriate amplitude and phase changes.

As r increases from O to $\pi/2$, the phase retardation Δ increases continuously. In any given direction the net state of polarization is obtained by adding the orthogonal states with their appropriate phase differences. In certain directions Δ will be integral multiples of 2π, i.e. the polarization of light on emergence is the same as that of the incident light. Between crossed polaroids light along such directions gets extinguished. Such a beam while traversing the medium traces out a closed loop on the Poincaré sphere. It is generally not recognized that along these directions, even in transparent optically inactive birefringent crystals, the emergent beam acquires the Pancharatnam phase[6]. This is due to phase on resolution, i.e. a phase arising from polarization of the incident light wave followed by analysation of the emergent light wave. It will depend upon the azimuth of the incident electric vector with respect to the major or the minor axis of the central elliptic section of the index ellipsoid for that particular direction of propagation.



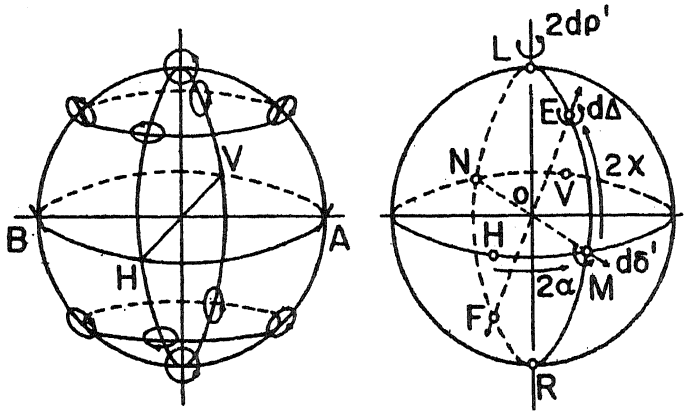
The phase on resolution is not obvious in the conoscopic patterns of optically inactive transparent birefringent crystals. However, there are instances where it is obvious in the pattern. In the case of optically active birefringent crystals (with or without absorption) like quartz or amethyst, between a circular polariser and a linear analyser, one finds a spiral interference figure in the conoscopic pattern. This spiral pattern was shown by Pancharatnam as arising from the excess phase on resolution. In other words, the pattern is a manifestation of phase on resolution.

Along any direction of propagation we combine the effects of linear dichroism and linear birefringence. Under their combined influence, the base states that travel unaltered are two similarly rotating (same handedness) elliptic vibrations with their major axes crossed and their ellipticities equal. In Figure 1 we have shown the procedure by which this answer can be obtained using the Poincaré sphere. Clearly these base

Box 2. The Poincaré sphere

In a completely polarized beam we require two parameters to describe a general state: (i) the azimuth λ, i.e. the orientation of the major axis of the ellipse, and (ii) the 'ellipticity', $\omega = \tan^{-1} b/a$, where a and b are the semi major and minor axes of the ellipse. Positive and negative ω's represent respectively the left and right rotating ellipses.

In 1892 Poincaré came up with an elegant method of representing a polarization state on the surface of a sphere by using 2ω and 2λ as the latitude and longitude. Figure *a* represents the Poincaré sphere. The poles L and R represent respectively the left and right circular vibrations and points on the equator *HAVB* represent linear vibrations. Points *H* and *V* are the horizontal and vertical linear vibrations. Every other point on the sphere represents an elliptic vibration.



a *b*

The Poincaré representation is ideally suited to deal with changes in the state of polarization of a beam of light traversing an anisotropic medium. Normally when there is no absorption the incident beam splits up into two orthogonal states represented by diametrically opposite points on the sphere. These are two linear in the case of a linear birefringence, two circular in the case of optical activity, and two orthogonal elliptic states in the case of birefringence and optical activity. The coherent superposition of these two orthogonal states gives points on a great circle joining them. Changing the relative phase is equivalent to the rotation of the incident state about the axis joining the two orthogonal states (see Figure *b*).

states are non-orthogonal. It is this non-orthogonality that results in an interference between them resulting in the fringe pattern in the Brewster's brushes and idiophanic rings. A study of this effect led Pancharatnam to develop his generalized theory of interference[6].

Optic axes. Along either of the optic axes the birefringence is zero, and hence the base states are X_k and Y_k, i.e. two linear orthogonal vibrations travelling with the same velocity but with different attenuations. As a consequence of this, any incident linearly polarized light, apart from losing some intensity, will also rotate towards the less absorbed vibration X_k. Thus between crossed polaroids, the light ray travelling along the optic axis will not get extinguished, a property reminiscent of optically active crystals.

Singular axes. In the neighbourhood of each of the two optic axes and in a plane perpendicular to the axial plane, there will be in general two directions along which $X_k Y_k$ and $X_r Y_r$ are at a relative angle of $\pm 45°$ (orthogonal to each other on the Poincaré sphere) with $\delta = \pm k$. From Figure 2 it can be easily seen that along such a direction, called a *Singular axis*, only the left (or right) circular state propagates unaltered. This brings us to an interesting question. What would happen if the orthogonal circular state falls on the crystal in the direction of the singular axis? Voigt[3], who was the first to point out the existence of singular axes, said that the orthogonal circular state would be unable to travel through the crystal and would be completely reflected back. Pancharatnam[4] gave the right answer to this question. It can be seen from Figure 2 that the orthogonal state C_r as it travels through the crystal, gradually goes towards the permitted state C_l along the meridional arc $C_r X_k C_l$. The states C_r and C_l represent right and left circular vibrations respectively. In this process it continuously undergoes a change in the polarization state which at any point in the medium is always closer

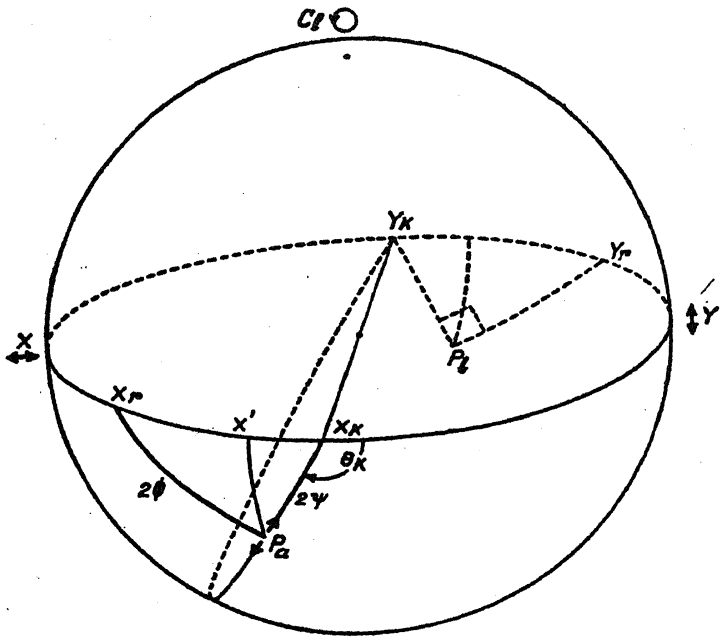


Figure 1. Poincaré sphere method[4] for working out the base states for any general direction of propagation in the case of an absorbing optically inactive biaxial crystal. X_r, Y_r and X_k, Y_k represent the principal planes of the elliptic sections of the index and absorption ellipsoids respectively. P_a and P_b are the base states.

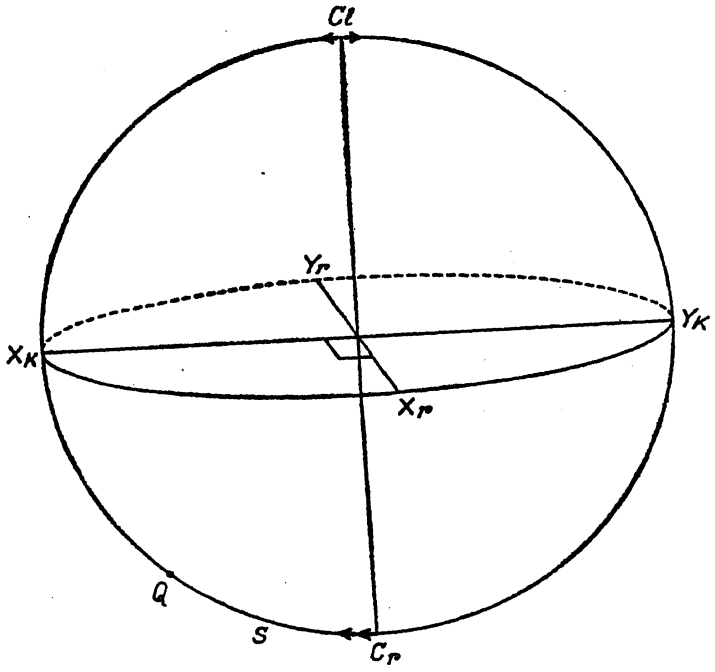


Figure 2. Poincaré sphere method[4] for obtaining the permitted state of propagation for the singular axis.

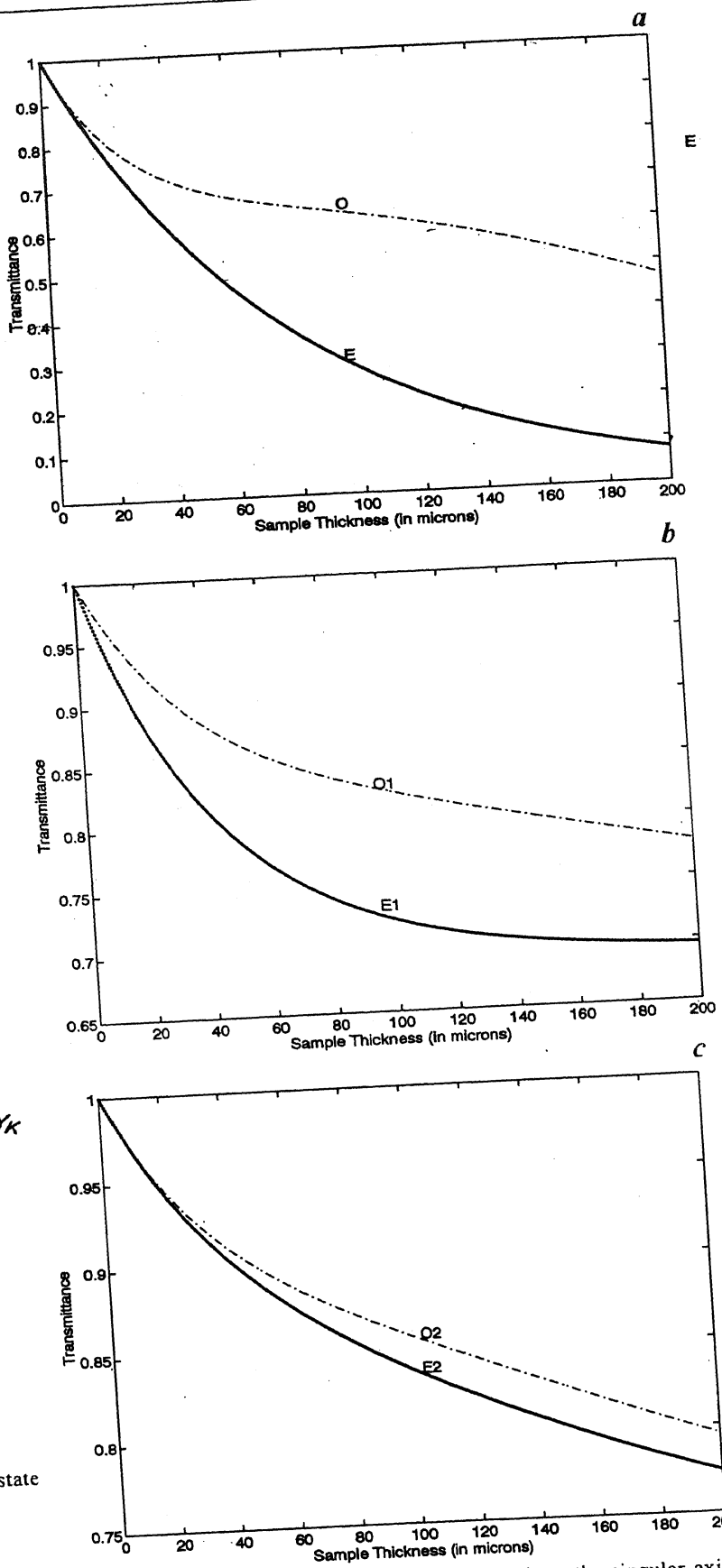


Figure 3. *a*, Transmittance for propagation along the singular axis The curve (E) is for the permitted eigenstate and (O) is for a stat with same azimuth and ellipticity but with opposite handedness. *b* and *c*, are for the two elliptic base states E_1 and E_2 permitted for an general direction of propagation. The state O_1 (O_2) is of sam azimuth and ellipticity as E_1 (E_2) but of opposite handedness.

to X_k (the less absorbed vibration) than C_l, the state that goes through unaltered. Hence it will always emerge with a relatively higher intensity. Pancharatnam not only predicted[4] this beautiful result but also confirmed it experimentally[7]. The effect can also be deduced from

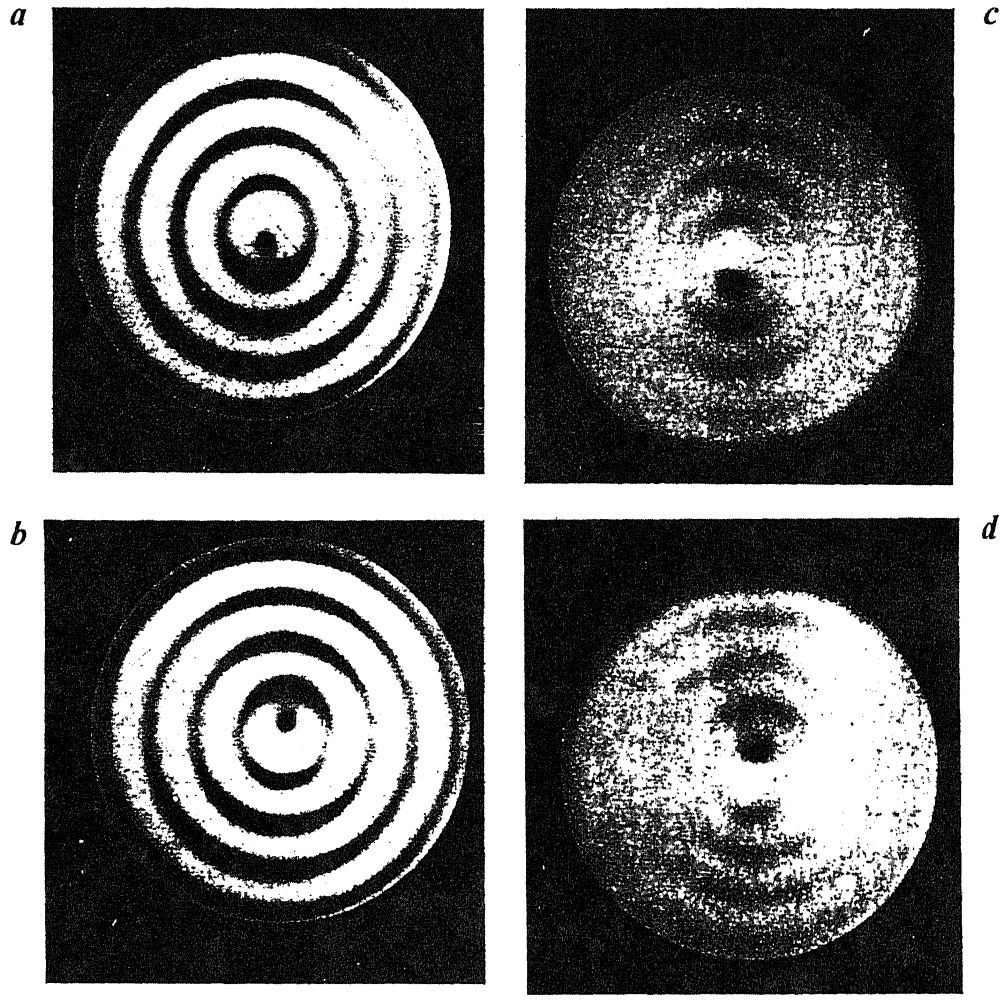


Figure 4. Conoscopic patterns in the neighbourhood of the singular axes[5]. The axial plane is horizontal. (*a*) Left circular polariser and right circular analyser. Note that the lower singular axis is extinguished; (*b*) Right circular polarizer and left circular analyser. Here the upper singular axis is extinguished; (*c*) Left circular polarizer alone. Note that the upper singular axis where the left circular vibration cannot be propagated unaltered actually appears brighter than the lower singular axis along which it can propagate unchanged; (*d*) Left circular analyser alone. Note that the lower singular axis appears brighter than the upper singular axis.

electromagnetic theory[8]. One such calculation is shown in Figure 3 *a*. It must be remarked that a somewhat similar phenomenon can exist in any general direction of propagation. A state that has the same azimuth and ellipticity as that of an eigenstate but of opposite handedness always emerges with a higher intensity compared to that state. This is shown in Figures 3 *b* and *c* for the two elliptic base states. The features associated with the singular axes are conspicuous in the conoscopic patterns shown in Figure 4.

Optically active crystals

Pancharatnam[9–11] also considered optically active absorbing crystals. The crystal of amethyst whose pleochroism he had once studied itself afforded an opportunity to verify his theoretical predictions. The important results in this case are:

(i) In any general direction two elliptic vibrations can travel unaltered. Their ellipticities, sense and azimuths do not bear any simple relation to each other.

(ii) Along the optic axes for $2\rho > k$, the base states are two elliptic vibrations exactly similar in form and orientation but described in opposite senses while for $2\rho < k$, these states are non-orthogonal linear vibrations, i.e. linear vibrations not at right angles to one another.

Highly pleochroic crystals

In the case of crystals with a large linear pleochroism and a comparatively small linear birefringence, the roles of k and δ get interchanged. Hence it is not near the optic axis that we find the singular axes, but near the direction of isotropic absorption (direction with $k = 0$).

Absorbing crystals in a magnetic field

The effects of an external magnetic field on the optical properties of an absorbing crystal can be worked out on

similar lines. Here we have to also consider the Faraday rotation ρ_f. We work out the properties for propagation along the optic axis.

Singular optic axis

We consider the case of an optically inactive absorbing crystal with a magnetic field along the optic axis. We have at low fields for which $2|\rho_f| < |k|$, two non-orthogonal linear vibrations as base states. At high fields for which $2|\rho_f| > |k|$, we get two elliptic vibrations of same form but of opposite senses as the base states. In addition, at the critical field H_c for which $2\rho_f = \pm k$, we find that for one direction of propagation only one linear state at + 45° to X_k can go through unaltered. While in the opposite direction it is the orthogonal linear vibration, that is the one at – 45° to X_k that can travel unaltered. Thus the optic axis becomes a singular axis. But this singular axis is quite different from what we find in field free absorbing crystals.

In the case of optically active absorbing crystals with negligible circular dichroism, for a field along an optic axis, in one direction the natural rotation ρ_n and the Faraday rotation ρ_f will add up and in the opposite direction they will oppose each other. Hence the net rotation ρ can be made equal to $\pm k/2$ for only one direction. Therefore the optic axis becomes singular only in one direction while in the opposite direction the base states corresponding to the $2\rho > k$ case will exist.

Elliptic dichroism

Pancharatnam[9] predicted that under the combined effects of linear and circular dichroisms, the medium becomes elliptically dichroic, i.e. two orthogonally polarized elliptic states travel unaltered with the same velocity but with different attenuation coefficients. This result can be directly established by studying light propagation along the optic axis of an absorbing optically active crystal. In the presence of a magnetic field, if the magneto-optic rotation ρ_f exactly annuls natural rotation ρ_n, then we end up with only linear and circular dichroisms along this direction and hence we get only elliptic dichroism.

Metals

Optics of anisotropic crystals

Metals have very high absorption for light waves. This is due to their free electrons. In spite of heavy optical absorption, in anisotropic metallic crystals we get interesting frequency-dependent effects. We know that a metal has a real refractive index for frequencies higher than the plasma frequency ω_P and it is completely imaginary for frequencies lower than ω_P. In an anisotropic metal for any direction of propagation there will be two plasma frequencies $\omega_{P_\parallel}$ and $\omega_{P_\perp}$ for the two principal vibrations. If the frequency ω of the incident vibration is higher than both $\omega_{P_\parallel}$ and $\omega_{P_\perp}$ the medium will be transparent. If ω is between $\omega_{P_\parallel}$ and $\omega_{P_\perp}$ then one principal vibration will be completely reflected and the other principal vibration will be completely transmitted. On the other hand, if ω is smaller than both $\omega_{P_\parallel}$ and $\omega_{P_\perp}$, then both the vibrations get reflected back. Under this category we can also consider 1D and 2D conductors. In such materials index and absorption tensor surfaces need not be closed surfaces. They could be even hyperboloids of one or two sheets[12]. Hence in biaxial highly anisotropic metals we need not have either the optic axes or the singular axes and the crystal will then exhibit many of the features associated with the $\omega_{P_\parallel} < \omega < \omega_{P_\perp}$ case.

Magneto-optics of isotropic metallic media

Cubic metals and the earth's ionosphere are optically isotropic in the absence of a magnetic field. However, we find interesting anisotropic properties in the presence of a magnetic field. The important results in this case are[13]:

(i) For propagation along the field direction the base states are right and left circular vibrations and the refractive indices of the medium to these base states are dependent on ω, the frequency of the light wave, ω_P the plasma frequency and ω_c the cyclotron frequency. At normal fields ω_c will be much less than ω and for metals ω_P will be generally in the violet or the ultraviolet end of the spectrum. At $\omega > \omega_P$, both the right and left circular vibrations travel with different velocities resulting in a Faraday rotation. In the narrow range of frequencies $(\omega_P - 1/2\ \omega_c) < \omega < (\omega_P + 1/2\ \omega_c)$ only the left circular vibration is a propagating wave and the right circular vibration becomes an evanescent wave. However, for $\omega < (\omega_P - 1/2\ \omega_c)$ both the circular states become evanescent.

(ii) For propagation perpendicular to the field we have two base states. The first is a transverse linear vibration parallel to the field and the second is an elliptic vibration in a plane perpendicular to the field. The second base state has both transverse and longitudinal components. The first base state is propagating only for $\omega > \omega_P$ and is an evanescent wave for $\omega < \omega_P$.

(iii) In the case of the ionosphere we find that the cyclotron frequency ω_c due to earth's magnetic field is higher than radio frequencies. Under these conditions the radio waves travel along the earth's magnetic field lines. These are called Whistler waves.

Spatial dispersion

Absorbing crystals are also good candidates for observing the effects of spatial dispersion. In the neighbourhood of its absorption bands, the refractive index of the medium increases and the absorption *per se* will not be very important for narrow absorption bands (this can happen at low temperatures). As the refractive index increases, the wavelength of light in the medium decreases. Under such conditions spatial dispersion leads to a dependence of the dielectric tensor on the wavevector $\boldsymbol{k}$ of propagation. The fact is that the familiar phenomenon of optical activity is itself a manifestation of spatial dispersion[14]. A careful analysis by Pekar[15] and Ginzburg[14] revealed many more interesting optical effects that can be attributed to spatial dispersion. We will emphasize only two of the important ones here.

Cubic crystals

The dielectric tensor ε_{ij} in a spatially dispersive non-dissipative medium is given by[14]

$$\varepsilon_{ij}(\omega k) = \varepsilon_{ij}(\omega) + i\,\gamma_{ijl}(\omega)\,k_l + \alpha_{ijlm}\,k_l\,k_m + \cdots$$

In a centrosymmetric system γ_{ijl} will not exist but α_{ijlm} will exist. In particular, α_{ijlm} will not vanish in cubic crystals resulting in a dependence of refractive index on the direction of propagation and polarization of the wave. Interestingly, not only does the cubic crystal become anisotropic, but it will also have seven optic axes, one each along the cube edge and the cube diagonal. It should be pointed out that due to spatial dispersion a cubic crystal also becomes pleochroic in its absorption bands.

Optically active crystals

In optically active crystals the base states are left and right crossed ellipses of same ellipticity. Interestingly spatial dispersion leads to slightly different ellipticities for these two vibrations. Another effect manifests itself when we go very close to any of its resonant frequency ω_o. Here

$$\varepsilon_{ij}(\omega) = \varepsilon(\omega) \approx \varepsilon_o - \frac{A\omega_o}{(\omega - \omega_o)} = \varepsilon_o - \frac{A}{\xi},$$

(neglecting absorption). In these crystals, for which $\gamma_{ijl} \neq 0$, one gets a dispersion of refractive index as shown in Figure 5. The two curves are for the two base states, viz. right and left circular vibrations. For one of the circular states we find a *turning point* at ξ_m. For $\xi < \xi_m$ we get two right (left) circular waves but travelling with different velocities and one left (right) circular wave. Thus we get a *new* circular wave. The two right (left) circular waves can interfere resulting in a thickness dependent transmitted intensity[13]. For $\xi > \xi_m$ we have only the left (right) wave propagating through the crystal. It may be remarked that a *new wave* is also found even in the optically inactive crystals near a resonant frequency. Spatial dispersion also leads to many other interesting properties[16].

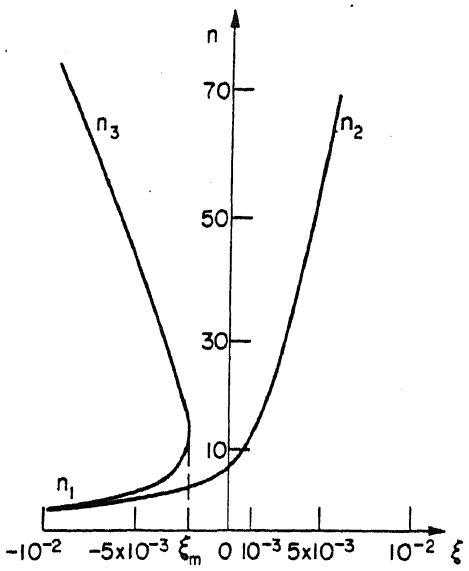


Figure 5. Dispersion curve in the resonance band of a spatially dispersing medium[13].

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