1.5.7.3. *Linear magnetic birefringence*

The magnetic contribution to the component of the dielectric permittivity $\delta\varepsilon_{ij}$ can be represented as a series in the powers of the components of the magnetization and the antiferromagnetic vector. The magnetic birefringence (also called the Cotton–Mouton or Voigt effect) is described by the real symmetrical part of the tensor $\delta\varepsilon_{ij}$. In paramagnetic crystals, the magnetization $\mathbf{M}$ is proportional to the applied magnetic field $\mathbf{H}$, and the series has the form

$$\delta\varepsilon_{ij} = Q^{MM}_{ijk\ell} M_k M_\ell = Q^{MM}_{ijk\ell}\chi^M_{kr}\chi^M_{\ell s} H_r H_s = \Gamma_{ijrs} H_r H_s. \quad (1.5.7.18)$$

The tensor Γ_{ijrs} is symmetric with respect to both the first and the second pair of indices. The symmetry of this tensor implies that the diagonal components of the permittivity tensor include magnetic corrections. The modification of the diagonal components gives rise to birefringence in cubic crystals and to a change $\Delta n^{\rm pm}$ of the birefringence in uniaxial and lower-symmetry crystals. It follows from (1.5.7.18) that this birefringence is bilinear in the applied field. Bilinear magnetic birefringence can be observed in uniaxial crystals if the magnetic field is applied along the x axis perpendicular to the principal z axis. In the simplest case, a difference in the refractive indices n_x and n_y arises:

$$\Delta n^{\rm pm} = n_x - n_y = \frac{1}{2n_0}(\delta\varepsilon_{xx} - \delta\varepsilon_{yy}) = \frac{1}{2n_0}(\Gamma_{xxxx} - \Gamma_{yyxx})H_x^2, \quad (1.5.7.19)$$

where n_0 is the refractive index for the ordinary beam.

Consider now a magnetically ordered crystal which can be characterized by an antiferromagnetic vector $\mathbf{L}_0$ and a magnetization vector $\mathbf{M}_0$ in the absence of a magnetic field. Applying a magnetic field with components H_r, we change the direction and size of $\mathbf{L}_0$ and $\mathbf{M}_0$, getting additional components $L^H_k = \chi^L_{kr}H_r$ and $M^H_k = \chi^M_{kr}H_r$. This is illustrated by the relations (1.5.7.6). Instead of (1.5.7.18) we get

$$\begin{aligned}\delta\varepsilon_{ij} &= Q^{LL}_{ijk\ell}L_kL_\ell + Q^{ML}_{ijk\ell}M_kL_\ell + Q^{MM}_{ijk\ell}M_kM_\ell\\ &= Q^{LL}_{ijk\ell}L_{0k}L_{0\ell} + Q^{ML}_{ijk\ell}M_{0k}L_{0\ell} + Q^{MM}_{ijk\ell}M_{0k}M_{0\ell}\\ &\quad + [2Q^{LL}_{ijk\ell}\chi^L_{kr}L_{0\ell} + Q^{ML}_{ijk\ell}(\chi^M_{kr}L_{0\ell} + M_{0k}\chi^L_{\ell r}) + 2Q^{MM}_{ijk\ell}\chi^M_{kr}M_{0\ell}]H_r.\end{aligned} \quad (1.5.7.20)$$

The terms in the middle line of (1.5.7.20) show that in an ordered state a change in the refractive indices occurs that is proportional to L_0^2 in antiferromagnets and to M_0^2 in ferromagnets. The terms in square brackets show that a linear magnetic birefringence may exist. In the special case of a tetragonal antiferromagnet belonging to the space group $\boldsymbol{D}^{14}_{4h} = P4_2/mnm$ with $\mathbf{L}_0$ parallel to the principal axis z, the linear birefringence occurs in the xy plane if the magnetic field is applied along the z axis (see Fig. 1.5.5.3). In this case, $\mathbf{M}_0 = 0$, $\chi^L_{kz} = 0$ for all k, $\chi^M_{xz} = \chi^M_{yz} = 0$ and $\chi^M_{zz} = 1/B$ [see (1.5.7.6)]. Therefore the terms in square brackets in (1.5.7.20) differ from zero only for one component of $\delta\varepsilon_{ij}$,

$$\delta\varepsilon_{ij} = Q^{ML}_{xyzz}L_{0z}H_z/B = q_{zxy}H_z\,{\rm sign}(L_{0z}). \quad (1.5.7.21)$$

As a result,

$$\Delta n^{\rm af} = n_{x'} - n_{y'} = \frac{1}{2n_0}\delta\varepsilon_{xy} = \frac{1}{2n_0}q_{zxy}H_z\,{\rm sign}(L_{0z}), \quad (1.5.7.22)$$

where x', y' are the optic axes, which in these tetragonal crystals are rotated by $\pi/4$ relative to the crystallographic axes.

Comparing relation (1.5.7.22) with (1.5.7.3), one can see that like LM, there may be linear magnetic birefringence. The forms of the tensors that describe the two effects are the same.

Linear magnetic birefringence has been observed in the uniaxial antiferromagnetic low-temperature α-Fe_2O_3 when the magnetic field was applied perpendicular to the threefold axis (Le Gall *et al.*, 1977; Merkulov *et al.*, 1981). The most impressive effect was observed in CoF_2 when the magnetic field was applied along the fourfold axis. The crystal ceased to be optically uniaxial and a difference $(n_{x'} - n_{y'}) \propto H_z$ was observed in accordance with (1.5.7.22). Such linear magnetic birefringence does not exist in the paramagnetic state. Linear birefringence has also been observed in $CoCO_3$ and $DyFeO_3$. For details of these experiments, see Eremenko *et al.* (1989). These authors also used linear birefringence to make the antiferromagnetic domains visible. A further review of linear magnetic birefringence has been given by Ferré & Gehring (1984).

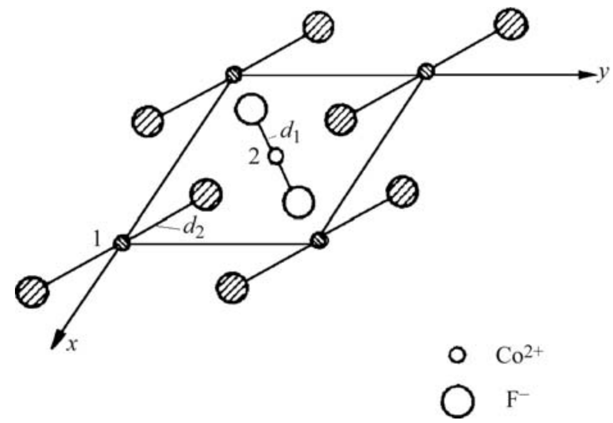

Fig. 1.5.7.3. Variation of symmetry of the crystal field in the presence of the piezomagnetic effect in CoF_2. The unshaded atoms lie at height $c/2$ above the xy plane (see Fig. 1.5.5.3).

Piezomagnetism, linear magnetostriction and linear birefringence in fluorides can be clearly demonstrated qualitatively for one particular geometry. As shown in Fig. 1.5.7.3, the crystallographically equivalent points 1 and 2 are no longer equivalent after a shear deformation applied in the plane xy. During such a deformation, the distances from the magnetic ions to the nearest fluoride ions increase in points 1 and decrease in points 2. As a result, the values of the g-factors for the ions change. Evidently, the changes of the values of the g-factors for different sublattices are opposite in sign. Thus the sublattice magnetizations are no longer equal, and a magnetic moment arises along the direction of sublattice magnetization. On the other hand, if we increase the magnetization of one sublattice and decrease the magnetization of the other by applying a magnetic field parallel to the z axis, the interactions with the neighbouring fluoride ions also undergo changes with opposite signs. This gives rise to the magnetostriction. These considerations can be applied only to antiferromagnets with the fluoride structure. In these structures, single-ion anisotropy is responsible for the weak ferromagnetism, not the antisymmetric exchange interaction of the form $\mathbf{d}[\mathbf{S}_i \times \mathbf{S}_k]$.

1.5.8. Magnetoelectric effect

Curie (1894) stated that materials that develop an electric polarization in a magnetic field or a magnetization in an electric field may exist. This prediction was given a more precise form by Landau & Lifshitz (1957), who considered the invariants in the expansion of the thermodynamic potential up to linear terms in H_i. For materials belonging to certain magnetic point groups, the thermodynamic potential Φ can be written in the form

$$\Phi = \Phi_0 - \alpha_{ij}E_iH_j. \quad (1.5.8.1)$$

If (in the absence of a magnetic field) an electric field $\mathbf{E}$ is applied to a crystal with potential (1.5.8.1), a magnetization will be produced:

$$M_j = -\frac{\partial\Phi}{\partial H_j} = \alpha_{ij}E_i. \quad (1.5.8.2)$$

  references