Bragg-regime diffraction and waveguiding of light by ferromagnetic domains in K_2CuF_4

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Bragg-regime diffraction by a thick magnetic phase grating is observed for the first time at perfectly transparent ferromagnetic stripe domains in K_2CuF_4 below $T_C = 6.2$ K in the visible region. The diffracted beams are perpendicularly polarized with respect to the incident and the transmitted beam. The diffraction vanishes by virtue of a new waveguiding mechanism owing to total reflection at the spatially thick, but optically thin, domain walls, if the incident light is polarized parallel to the crystalline *a* axis.

Thick dielectric phase gratings, which obey Bragg's law and exhibit only one single diffracted and one transmitted beam play a fundamental role in applied wave physics, e.g., in optical holography. This is well documented in a large number of publications, which appeared in the last 15 years. On the other hand only very little is known of analogous thick magnetic gratings despite their obvious practical interest. They would be very welcome, e.g., for magnetically controlled, highly efficient light deflection systems. However, since the pioneering work of Dillon and Remeika[1] and Lambeck[2] without exception only thin magnetic gratings have been observed and theoretically treated.[3] In contrast with thick gratings one finds higher-order diffraction maxima, the intensity of which is determined by the object phase function. The three-dimensional nature of the grating can virtually be neglected.

In this Report we shall present data on the two-dimensional (2D) ferromagnet K_2CuF_4, which for the first time offers the opportunity to study magnetic light diffraction in the Bragg regime. K_2CuF_4 is perfectly transparent in the visible region, unlike most of the materials investigated before,[3] and it presents a regular ferromagnetic stripe domain structure below its Curie temperature $T_C = 6.2$ K.[4] The domains lie perpendicular to the tetragonal *c* axis, parallel to the easy plane of magnetization. When viewed along one of the *a* axes, a sample cut parallel to an *ac* face obtains the phase structure of a typical magnetic grating. It is due to the magneto-optical Faraday effect coupled to the alternating longitudinal magnetization M within adjacent domains. The direction of M is fixed parallel to [110] and [$\bar{1}$10], respectively, by a very small, but finite intraplanar anisotropy field, $H_A^{in} = 0.1$ Oe.[4] Its smallness also explains the exceptionally large wall width, $W' \sim 1$ μm, which is nearly comparable with that of the domains, $W \sim 3$ μm. Owing to the rotation of M within the Bloch walls and to the closure domains having M parallel to the sample surface,[4] the modulation of the circular refractive index has a triangular rather than the usually observed[3] square shape.

The microscopic image of the domain structure of a typical sample (thickness $L = 1.5$ mm), placed between crossed polarizers and recorded at 4.8 K with $\lambda = 546$ nm light, is shown in Fig. 1(a). In order to eliminate the strong tetragonal birefringence in the *ac* plane[4] we can choose between two configurations, the E and the H mode,[5] i.e., the $\vec{E}$ vector of the incident light being parallel and perpendicular, respectively, to the grating vector $\vec{K} \parallel \vec{c}$. Obviously only in the E mode the periodicity of the phase object becomes visible due to the Faraday contrast between bright bulk domains and dark walls. In the H mode the image is structureless and consists of diffuse bright stripes without any distinct periodicity.

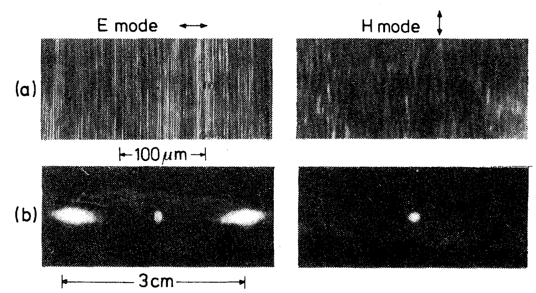

FIG. 1. Microscopic images (a) and diffraction patterns at a distance of 0.2 m (b) obtained at 4.8 K with 546-nm light on an *ac* section of K_2CuF_4 placed between crossed polarizers. The polarization of the incident light is indicated by arrows.

As a consequence and in agreement with Abbe's theory of the microscopic image, the Fraunhofer diffraction patterns are quite different in both modes. In a classical large-aperture diffraction setup outside the microscope we find first-order spots in the Fourier plane for the E mode, but no diffracted beam at all in the H mode at normal incidence [Fig. 1(b)]. In both modes the transmitted zero-order beams are nearly extinguished by the crossed analyzer, hence conserving essentially the original polarization. The diffracted beams in the E mode, on the other hand, are perfectly perpendicularly polarized with respect to the incident polarization. Neither the diffraction, nor the image patterns change qualitatively at different $L\,(0.8-4.5$ mm) or $\lambda(400-650$ nm) or, if a magnetic field parallel to the stripes being smaller than the collapse field $H_c \sim 100$ Oe is applied.[4] Details are planned to be given elsewhere.[6]

Discussing at first the diffraction in the E mode, we remark that the diffraction spots are larger than the image of the entrance pupil appearing in the zero-order maximum. This is due to local variations of the lattice constant, which are also visible in the image. Virgin domain structures generally yield even broader diffraction spots, which can be reduced to the size shown in Fig. 1(b) by aligning the domains with a small magnetic field[4] prior to the measurement.

The first-order diffraction angle outside the crystal is given by $\sin\vartheta = (\lambda/D)(n_a/n_c)$, where λ/n_c is the wavelength of the incident light inside the crystal, which is transformed into a-polarized light after diffraction. Hence, the refraction at the rear side of the crystal implies $\sin\vartheta = n_a \sin\vartheta'$, where ϑ' is the diffraction angle inside the sample. Using the experimental data of Fig. 1(b), $\vartheta = 4.1°$ and $\lambda = 546$ nm, and taking $n_a/n_c \sim 1$,[4] we obtain the grating constant $D = W_+ + W_- + 2W' = (8.2 \pm 1.6)$ μm in agreement with the microscopic image [Fig. 1(a)] and previous results.[4] W_+ and W_- are the widths of up and down magnetized adjacent bulk domains, respectively. The standard deviation $\Delta D = \pm 1.6$ μm is estimated from the widths of the diffraction spots.

The complete absence of all but the first-order diffraction maxima is a first hint at a diffraction mechanism of the Bragg-reflection type. We find neither third-order maxima, which have about $\frac{1}{10}$ of the first-order intensity in thin square magnetic gratings,[3] nor second-order spots, even when using unequally spaced domain configurations,[3] stabilized by an external magnetic field.[4] K_2CuF_4 thus differs in a fundamental way from all heretofore-investigated systems.[3] Most convincing evidence, however, for the Bragg-like diffraction is derived from its angular dependence. Very typically of thick gratings the diffraction pattern becomes asymmetric at nonzero angle of incidence θ [Fig. 2(a)]. Simultaneously the intensity concentrates on the outward moving spot ($m = +1$), attaining its maximum at $\theta_0 \sim 2°$, whereas

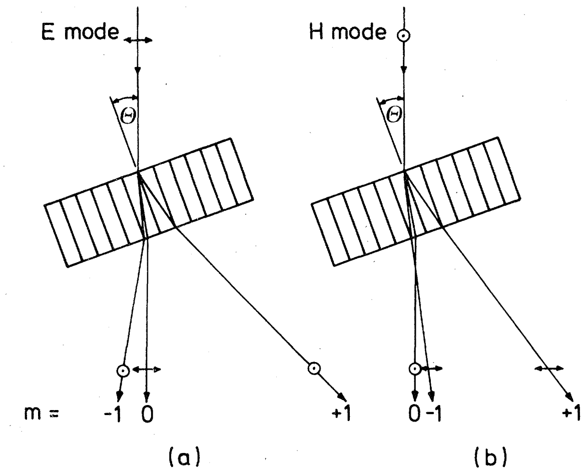

FIG. 2. Schematic view of the diffraction at inclined domain gratings of K_2CuF_4 for different light polarization as indicated by arrows.

the inward moving spot ($m = -1$) gradually weakens ($I_{+1}/I_{-1} \sim 20$ at θ_0). Obviously θ_0 corresponds to the internal Bragg angle $\theta_0' = \sin^{-1}[\lambda/(2n_c D)]$ of the $m = +1$ beam. With $\sin\theta_0 = n_c \sin\theta_0'$, $\lambda = 546$ nm, $n_c = 1.6$,[7] and $D = 8.2$ μm we calculate $\theta_0 = 1.9°$. It is well known[5] that the Bragg condition is not absolutely sharp for finite L. Hence, owing to the small θ_0, even at normal incidence one works near the Bragg angle and obtains symmetric, but relatively weak first-order maxima [Fig. 1(b)].

Quantitative aspects of the angular dependence of the diffracted intensity distribution will be given in our future paper,[6] which will comprise a multiwave theory of thick magnetic gratings along the lines worked out for thick dielectric gratings.[5,8,9] This theory contains the case $L \rightarrow 0$ as treated by Kuhlow and Lambeck[3] and explains the polarization rules found experimentally. Here we shall confine ourselves to note the criteria for Bragg-regime diffraction of magnetic gratings. Similarly to those of linear index gratings,[10] one needs simultaneously a large thickness parameter, $Q = (2\pi L\lambda)/(n_c D^2 \cos\theta)$ and a small modulation parameter, which for a magnetic gratings reads $\gamma = \beta/(2\cos\theta)$, where the Faraday rotation angle β stands for the familiar index modulation $\Delta n = (n_+ - n_-)/2 = (\lambda\beta)/(2\pi L)$. Inserting our experimental parameters ($L = 1.5$ mm, $n_c = 1.6$, $D = 8$ μm, $\beta = 8°$,[4] $\lambda = 546$ nm, $\theta = 0$) we obtain $Q = 48$ and $\gamma = 0.07$, hence $\rho \equiv Q/2\gamma = 343$. Supposing that the border line between normal and Bragg-regime diffraction is marked by a critical value $\rho_c \sim 10$ as in the case of dielectric gratings,[10] it is now formally confirmed that the stripe domains in K_2CuF_4 have to be treated as a "thick" grating. They thus represent the first elementary example of a "magnetic volume hologram."

It may be noted that the "thick" $CrBr_3$ samples studied by Kuhlow and Lambeck[11] do not fulfill the thickness conditions in the sense defined above. Based on their experimental data ($L = 20$ μm, $n = 2.73$, $D = 2.5$ μm, $\beta = 78°$, $\lambda = 546$ nm, $\theta = 0$) we calculate $Q = 4.0$ and $\gamma = 1.4$, hence $\rho = 1.4$, which lies near the upper bound of the normal, but not yet in the Bragg regime. Owing to its large Faraday rotation $CrBr_3$ exhibits multiple diffraction as a second-order effect, which leads to even-order diffraction maxima also for equally spaced domains.[11] Obviously this effect needs not be considered for K_2CuF_4.

The absence of any diffraction in the H mode at normal incidence (Fig. 1) is not expected either in thin,[3] or in thick purely magnetic gratings.[6] It must, hence, be due to the linear refractive-index modulation within domains and walls, which has recently been calculated for K_2CuF_4 (Ref. 12):

$$n_a(\alpha) = n_{a0} - n_{a0}^3 M^2[\rho_{11} + \rho_{12} + (\rho_{11} - \rho_{12})\cos 2\alpha]/2 \ , \tag{1}$$

$$n_c(\alpha) = n_{c0} - n_{c0}^3 M^2 \rho_{31}/2 \ , \tag{2}$$

where n_{a0}, n_{c0} are the unperturbed indices and α denotes the angle between M and the a axis parallel to the grating surface. ρ_{11}, ρ_{12}, ρ_{31} are magneto-optical constants, the microscopical origin of which has been discussed elsewhere.[12] The H-mode anomaly may be anticipated remarking that only n_a depends on α. As shown in Fig. 3, n_a is intermediate in the domains ($\alpha = \pm\pi/4$), whereas the extreme values at $\alpha = 0, \pi/2, \pi$ [Eq. (1)] are attained within the Bloch walls. Assuming $\rho_{11} > \rho_{12}$ and taking into account that $n_a(\alpha = 0;\pi)$ becomes largely enhanced because of the superimposed Faraday rotation[12] we arrive at the asymmetric index profile shown in Fig. 3, the values being valid for $\lambda \sim 550$ nm.[12]

In principle the index modulation forms a grating with a constant $D/2$, which should yield Bragg-like first-order maxima at the positions of the second-order magnetic diffraction spots. The negative experimental result, however, hints at a mechanism, which simultaneously suppresses both types of diffraction. In our opinion, this must be the total reflection at the domain walls, which by chance fulfill two necessary requirements: (i) they are optically thin with respect to the domains; and (ii) they are thick enough to damp out the penetrating attenuated wave. Since the latter condition is not met in strongly anisotropic systems,[3] the peculiar transformation of the domains into a system of uncoupled waveguides has never before been observed. Since the sample is not interferometrically flat, diffraction at its rear surface will

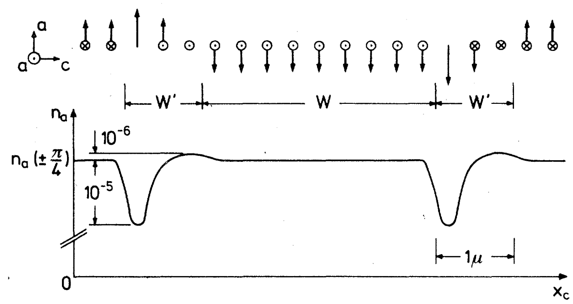

FIG. 3. Refractive index n_a as a function of the distance x_c along the c direction in the ac plane of K_2CuF_4. The direction of M within domains (width W) and walls (W') is presented schematically in the upper part (side-on view).

be negligible as well. The aperture angle of the flat waveguides obeys the relation

$$\sin\theta_{\max} = [n_a^2(\pi/4) - n_a^2(\pi)]^{1/2} \ , \tag{3}$$

which already accounts for the refraction at the surface and yields $\theta_{\max} = 0.4°$ using $n_{a0} = 1.6$ and $n_a(\pi/4) - n_a(\pi) = 10^{-5}$. This limit becomes slightly larger owing to the closure domains[4] acting as input prisms for the somewhat less refracting bulk domains.[12]

At oblique incidence, $\theta \neq 0$, indeed H-mode diffraction arises, however, not abruptly at $\theta \sim \theta_{\max}$, but only smoothly at $\theta > 3°$. This discrepancy is probably due to partial waveguiding along the grating at small angles, where the reflection coefficient is still large, although smaller than unity. In contrast with the E mode [Fig. 2(a)] both diffracted beams, $m = \pm 1$, appear at the same side with respect to $m = 0$ [Fig. 2(b)]. This peculiarity is due to the birefringence ($n_a > n_c$), since the emerging beams are differently polarized. It is also observed on samples with misoriented (that is, slanted) domains at normal incidence, which is planned to be discussed in detail elsewhere.[6]

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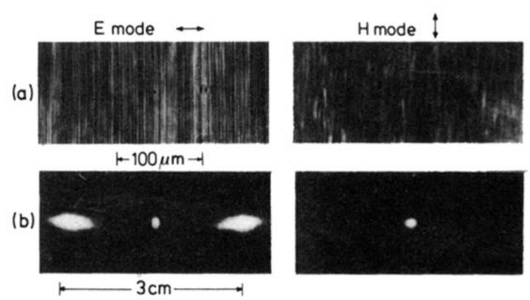

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