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Negative refraction in natural ferromagnetic metals

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Abstract – It is generally believed that Veselago's criterion for negative refraction cannot be fulfilled in natural materials. However, considering imaginary parts of the permittivity (ε) and permeability (μ) and for metals at not too high frequencies the general condition for negative refraction becomes extremely simple: $\mathrm{Re}(\mu) < 0 \Rightarrow \mathrm{Re}(n) < 0$ and may be fulfilled for such natural ferromagnetic metals as nickel, iron, or cobalt. Here we demonstrate experimentally that in pure cobalt and Fe/Co alloy the negative values of the refractive index can indeed be achieved close to the frequency of the ferromagnetic resonance. Large values of the negative refraction can be obtained at room temperature and they can easily be tuned in moderate magnetic fields.

Introduction. – The topic of negative refraction [1] has attracted much research interest in the last years [2,3]. Various possible realizations of a negative index material have been proposed [4–10]. Experimental demonstrations of negative refraction in metamaterials [11,12], multilayers [7] and ferromagnetic wires [13] are based on the classical criterion by Veselago which requires simultaneous negativity of electric permittivity ε and magnetic permeability μ [1]. However, natural materials with such exotic properties most probably do not exist. An alternative way to achieve negative refraction $\mathrm{Re}(n) < 0$ in natural materials may try to use an extended criterion which takes into account the imaginary parts of the permittivity, $\varepsilon_1 + i\varepsilon_2$ and permeability, $\mu_1 + i\mu_2$. Various equivalent forms of the extended criterion have been presented and detailed derivation of the corresponding expression has been given [14–17]. The starting point of such considerations is the condition

$$\mathrm{Re}(n) = \mathrm{Re}(\sqrt{\varepsilon\mu}) < 0. \tag{1}$$

For passive materials the energy conservation requires in addition that $\mathrm{Im}(n) > 0$. These two conditions now directly lead to the relation $\mathrm{Im}(\varepsilon\mu) < 0$, and finally, to

$$\frac{\mu_1}{\mu_2} + \frac{\varepsilon_1}{\varepsilon_2} < 0. \tag{2}$$

Here $\varepsilon = \varepsilon_1 + i\varepsilon_2$ and $\mu = \mu_1 + i\mu_2$ denote the complex dielectric permittivity and complex magnetic permeability, respectively.

Equation (2) has an interesting consequence if we consider the electrodynamics of metals at low frequencies [18]. For metals at frequencies far below the scattering rate (*i.e* in the Hagen-Rubens limit) the imaginary part of the permittivity dominates: $|\varepsilon_1| \ll \varepsilon_2$ and, therefore, $\varepsilon_1/\varepsilon_2 \approx 0$. In this case eq. (2) can be simplified even more:

$$\mu_1 < 0 \Rightarrow \mathrm{Re}(n) < 0. \tag{3}$$

The condition of eq. (3) may be fulfilled in ferromagnetic metals close to the ferromagnetic resonance if the strength of the mode is high enough. First experimental demonstration of a negative refraction in a ferromagnetic metal [9] utilized the ferromagnetic resonance in a colossal magnetoresistance manganite (La,Ca)MnO_3. Although a sufficiency of the simple condition $\mu_1 < 0$ for metals has been proven, it still remains to be shown that eq. (3) works for such classical metals like Fe, Ni, or Co.

In this paper we present the results of the millimeter wave transmission experiments on the refractive index in real natural metals. As examples of natural ferromagnetic metals we have chosen pure cobalt and Fe/Co alloy. Using polarization and phase controlled experiments we demonstrate that close to the frequency of the ferromagnetic

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resonance the refractive index of Co and Fe/Co alloy indeed goes deep into the negative regime.

Experimental details. – Pure cobalt and $Fe_{0.5}/Co_{0.5}$ alloy were prepared as polycrystalline thin films with an overall size of 10 mm × 10 mm by magnetron sputtering technique. The thickness of both samples was 150 ± 30 nm. As a substrate MgO was used, whose millimeter wave properties have been determined in a separate experiment as $n_{\mathrm{MgO}} = [3.09 + 1.4 \cdot 10^{-9}(T\,[\mathrm{K}])^3 \pm 0.01]$. Here the temperature T is given in kelvin. The imaginary part of the refractive index of the MgO substrate in the frequency range of the present experiment was below $1 \cdot 10^{-4}$ at all temperatures and could be neglected.

The experiments in the millimeter frequency range were carried out in a Mach-Zehnder interferometer setup [19,20] using backward wave oscillators as radiation sources. This spectrometer enables to measure both the transmittance and phase shift as function of frequency, temperature or external applied magnetic field within controlled polarization geometries. All measurements have been done in a normal incidence geometry. As the wavelength of the radiation within the sample is much smaller than the lateral dimensions of the sample, the diffraction effects could be neglected in the considerations. Magnetic field experiments were carried out using a split coil magnet with polypropylene windows.

The experimental spectra obtained were analyzed using the Fresnel formulas for the complex transmission coefficient of the substrate-film system [19–21]. In a simple form which neglects the substrate these equation may be written as

$$t = \frac{(1-r^2)\,t_1}{1-r^2 t_1^2}, \tag{4}$$

where $r = \frac{\sqrt{\varepsilon/\mu}-1}{\sqrt{\varepsilon/\mu}+1}$ and $t_1 = \exp(-2\pi i\sqrt{\varepsilon\mu}d/\lambda)$. Here r describes the reflection amplitude at the air-sample interface, t_1 is the "pure" transmission amplitude, d the film thickness and λ the wavelength of the radiation. Equation (4) can be easily extended to include the substrate properties [21,22].

In present experiments, we obtained the transmittance and phase shift of the sample within two different polarizations of the incident radiation, $\tilde{h}||B$ and $\tilde{h} \perp B$. Here $\tilde{h}$ is the ac magnetic field of the radiation and B is the external magnetic field. From the solution of Bloch's equations for the magnetic moments in external magnetic fields it follows that nonzero magnetic susceptibility can only be obtained in the geometry $\tilde{h} \perp B$ [23]. The inset of fig. 1 shows the geometry in which the ferromagnetic resonance is excited. To determine the parameters of the ferromagnetic resonance a Lorentz line shape was used, *i.e.* the magnetic permeability was taken as

$$\mu(B) = 1 + \frac{\Delta\mu B B_0}{B^2 - B_0^2 - iB_0\Gamma}. \tag{5}$$

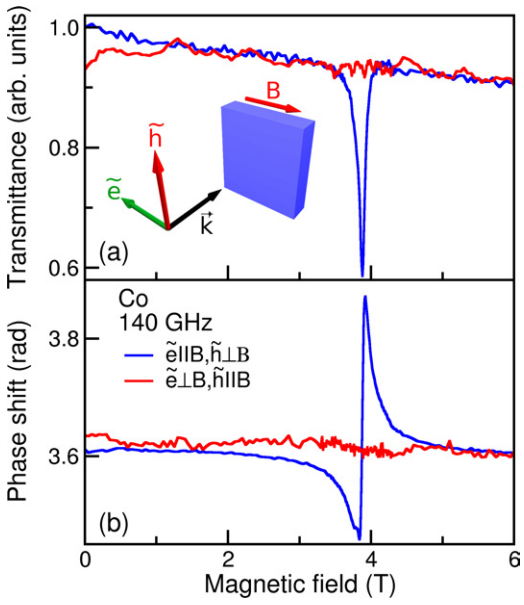

Fig. 1: (Colour on-line) Relative transmittance (a) and phase shift (b) of the metallic Co film for two different excitation geometries at $T = 190$ K. Within the geometry $\tilde{h} \perp B$ the ferromagnetic resonance can be excited, whereas for $\tilde{h}||B$ no excitation occurs. The inset sketches the magnetically active geometry ($\tilde{h} \perp B$). Here $\bar{k}$ denotes the wave vector, $\tilde{h}$ and $\tilde{e}$ are the ac magnetic and electric fields of the radiation, and B_0 is the static external magnetic field.

Here Γ is the width of the resonance, B_0 the resonance field and B the external applied magnetic field.

Figure 1 shows the transmittance and phase shift of the Co sample at $T = 190$ K and at a frequency of 140 GHz in both excitation geometries. As can be clearly seen in the geometry $\tilde{h} \parallel B$ no excitation of the magnetic resonance occurs (*i.e.* $\mu = 1$). Furthermore, in this geometry no magnetoresistance could be observed within our experimental resolution neither in the pure Co sample nor in the Fe/Co alloy. Therefore we can assume a magnetic-field–independent behavior of the permittivity. The dielectric properties of the metal films are now obtained from the absolute transmittance and phase shift spectra in zero magnetic field, which are shown in fig. 2. These spectra can be well described using the Drude conductivity of a metal in the zero-frequency limit,

$$\sigma(\omega) = \sigma_{dc} - i\varepsilon_0\varepsilon_\infty\omega. \tag{6}$$

Here ε_0 is the permittivity of vacuum and ε_∞ is the dielectric contribution from the high-frequency processes (*e.g.* phonons). In a good approximation both σ_{dc} and ε_∞ can be taken as frequency independent in the millimeter wave range. The dielectric permittivity of metal is given by

$$\varepsilon_1 = \mathrm{Re}(\varepsilon) = \varepsilon_\infty, \tag{7}$$

$$\varepsilon_2 = \mathrm{Im}(\varepsilon) = \frac{\sigma_{dc}}{\varepsilon_0\omega}. \tag{8}$$

Table 1: Electrodynamic parameters for Fe/Co and Co samples at two different excitation frequencies and temperatures. $B(1,2)$, $\Delta\mu(1,2)$ and $\Gamma(1,2)$ are the resonance fields, magnetic contributions, and widths of the ferromagnetic resonances, respectively.

Sample	Fe/Co		Co	
frequency [GHz]	140	182	140	180
$T = 10\,\mathrm{K}$				
$B(1)$ [T]	3.73 ± 0.01	5.11 ± 0.01	3.87 ± 0.01	5.20 ± 0.01
$\Delta\mu(1)$	0.34 ± 0.09	0.18 ± 0.04	0.20 ± 0.04	0.12 ± 0.03
$\Gamma(1)$ [T]	0.07 ± 0.02	0.07 ± 0.02	0.08 ± 0.02	0.11 ± 0.02
$B(2)$ [T]	-	5.04 ± 0.01	3.78 ± 0.01	5.11 ± 0.01
$\Delta\mu(2)$	-	0.20 ± 0.09	0.11 ± 0.02	0.03 ± 0.02
$\Gamma(2)$ [T]	-	0.14 ± 0.02	0.10 ± 0.02	0.11 ± 0.02
$\sigma_{dc}[10^6\,\Omega^{-1}\mathrm{m}^{-1}]$	8 ± 2		12 ± 2	
$\varepsilon_1[10^5]$	-0.3 ± 2.0	-0.3 ± 2.0	-0.5 ± 2.0	-0.5 ± 2.0
$\varepsilon_2[10^5]$	10.8 ± 2.0	8.4 ± 2.0	15.4 ± 2.0	12.0 ± 2.0
$T = 300\,\mathrm{K}$				
$B(1)$ [T]	3.76 ± 0.01	5.17 ± 0.01	3.89 ± 0.01	5.21 ± 0.01
$\Delta\mu(1)$	0.25 ± 0.08	0.32 ± 0.08	0.38 ± 0.09	0.22 ± 0.08
$\Gamma(1)$ [T]	0.08 ± 0.02	0.10 ± 0.02	0.07 ± 0.02	0.12 ± 0.02
$B(2)$ [T]	3.66 ± 0.01	5.07 ± 0.01	3.79 ± 0.01	-
$\Delta\mu(2)$	0.10 ± 0.05	0.14 ± 0.07	0.12 ± 0.07	-
$\Gamma(2)$ [T]	0.11 ± 0.02	0.09 ± 0.02	0.11 ± 0.02	-
$\sigma_{dc}[10^6\,\Omega^{-1}\mathrm{m}^{-1}]$	6 ± 1		6 ± 1	
$\varepsilon_1[10^5]$	-0.2 ± 2.0	-0.2 ± 2.0	-0.2 ± 2.0	-0.2 ± 2.0
$\varepsilon_2[10^5]$	7.7 ± 2.0	5.9 ± 2.0	7.1 ± 2.0	5.5 ± 2.0

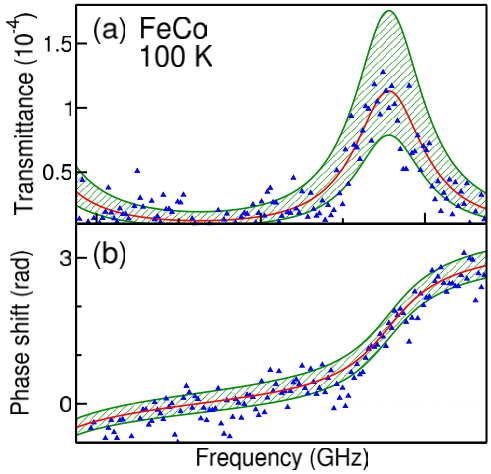

Fig. 2: (Colour on-line) Example of the transmittance (a) and phase shift (b) spectra (symbols) for a Fe/Co film at 100 K and at zero magnetic field (symbols). Solid lines correspond to fits using a Drude model, eq. (6), with $\sigma_{dc} = (10 \pm 2) \cdot 10^6\,\Omega^{-1}\mathrm{m}^{-1}$ and $\varepsilon_\infty = (-0.4 \pm 2.0) \cdot 10^5$. The shaded area denotes the 67% confidence interval of the fit.

The 67% confidence interval is shown from which the errors shown in table 1 are obtained. We note that in the case of a metal in the low-frequency limit the uncertainty of the transmittance mainly influences the conductivity and the uncertainty in the phase shift is basically responsible for the errors in $\mathrm{Re}(\varepsilon)$. From the spectra in fig. 2 we obtain the background value of the dielectric permittivity by numerical inversion of Fresnel equations. The values for the complex permittivity together with the fit values of the Drude conductivity are summarized in table 1.

In the geometry $\tilde{h} \perp B$ the ferromagnetic resonance can be excited which allows to independently determine the permeability as a function of the external magnetic field. Here we note that in the experimentally used geometry (Voigt-geometry) no off-diagonal elements in the permeability tensor are introduced [24], so μ takes the form

$$\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \mu(B) \end{pmatrix}. \tag{9}$$

Since the measurements were done in normal incidence and with radiation polarized along the magnetic axes of the sample, we may use the isotropic form of the Fresnel equations shown in eq. (4). We note that the negative refraction effect is not isotropic and holds solely for this experimental geometry. The resonance frequency is dependent on the external magnetic field and follows the relationship $\omega_{res} = \gamma\sqrt{(H_0 + 4\pi M_0)H_0}$ in the geometry of our experiment (Voigt geometry) [25]. Here ω_{res} denotes the frequency of the ferromagnetic resonance, γ is the gyromagnetic ratio and M_0 is the static magnetization. Using the dielectric permittivity obtained in zero magnetic field we are able to calculate the magnetic permeability as a function of the external field, which is shown in fig. 3 for the Co sample at $T = 190\,\mathrm{K}$. We note that the second weak

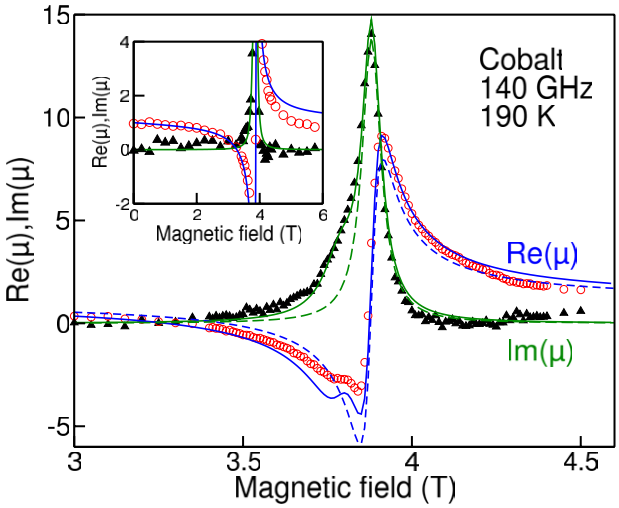

Fig. 3: (Colour on-line) Complex magnetic permeability of Co film as obtained from transmission and phase shift data (symbols) and fitted using eq. (5) (solid lines). Close to the resonance the real part of the permeability becomes negative. Dashed lines indicate a fit with a single resonance in μ. The inset shows the full range of the magnetic field scan.

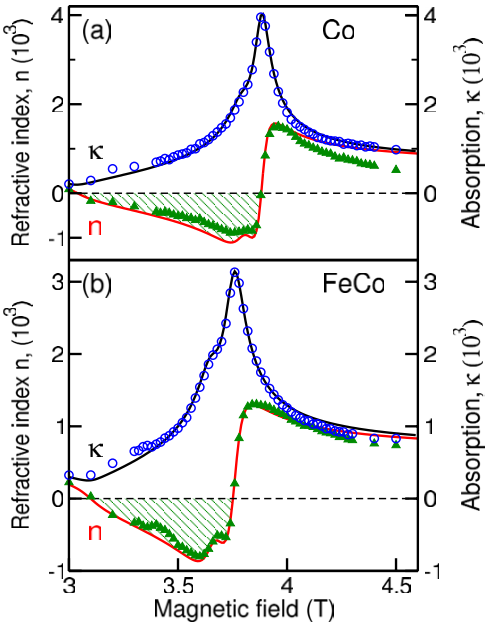

Fig. 4: (Colour on-line) Magnetic field dependence of the refractive index in (a) metallic Co and (b) Fe/Co alloy at $T = 300\,\mathrm{K}$ and at 140 GHz. The shaded area denotes the range in which the real part of the refractive index becomes negative. Symbols are for experimental data, solid lines are fits using eq. (5) and the parameters from table 1.

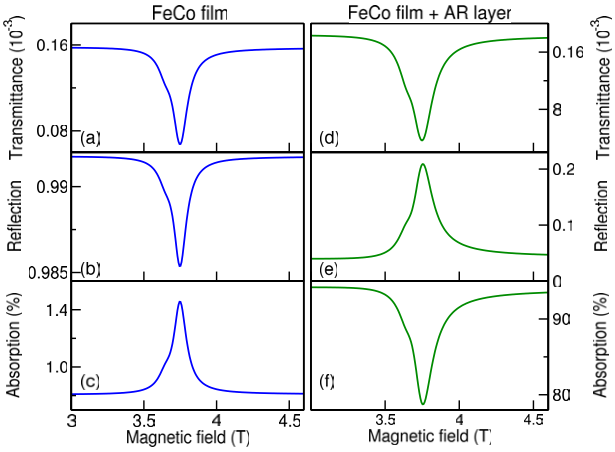

Fig. 5: (Colour on-line) Magnetic field dependence of (a) transmission, (b) reflection and (c) absorption for the Fe/Co sample determined from the fits to the complex transmission coefficient. (d)–(f) The same field scans for the Fe/Co layer with additional anti-reflection layer ($n_{AR} = 27$, $d_{AR} = 20\,\mu$m).

resonance which can be seen in these plots (at $\sim 3.7\,\mathrm{T}$ for Fe/Co and $\sim 3.8\,\mathrm{T}$ for Co) most probably arises due to inhomogeneities in the film magnetization [26]. In addition in fig. 3 also fits with a single resonance in μ are shown, which differ significantly from the experimental observed values. This can also be seen in the transmission and phase shift data (not shown).

Results and discussion. – Table 1 summarizes the electrodynamic films properties for Fe/Co and Co at two different excitation frequencies. For both samples we get quite similar electrodynamic parameters. Weak temperature dependence of the resonance fields is due to the decreasing static magnetization with increasing temperature. These effects are quite small, since the Curie temperatures of both Co and Fe/Co are far above room temperature (Co: $T_C = 1390\,\mathrm{K}$ [27], Fe/Co: $T_C = 1250\,\mathrm{K}$ [28]).

With the dielectric permittivity and the magnetic permeability determined, we can now calculate the refractive index by $\mathrm{n} = \sqrt{\varepsilon\mu}$. Figure 4(a) and (b) show the magnetic field dependence of the complex refractive index of the Fe/Co and Co sample, respectively. The shaded area denotes the range in which the refractive index is negative. Here we see that even at room temperature these metals show a negative index of refraction, which is also highly tunable by the applied magnetic field. The estimate of the best value of the figure of merit for our samples gives $|n|/\kappa \sim 0.8$. This value is already close to the theoretical limit $|n|/\kappa = 1$ for ferromagnetic metals.

Although the refractive index of Fe and Fe/Co films can be negative, the transmittance of our samples remains very low ($\sim 10^{-4}$). There are two main reasons which lead to the low values of the transmitted signal. On the one hand the absorption coefficient of these ferromagnetic metals is high in the region of negative refraction (fig. 4). On the other hand the reflectivity of metals in general is also quite high. This can be seen in fig. 5 which shows the magnetic field dependent transmission (a), reflection (b) and absorption (c) for the Fe/Co sample determined by the Fresnel equations using the fits for the permeability following eq. (5) and the parameters from table 1. At the

ferromagnetic resonance the overall absorption does not exceed 1.5%, since the most intensity becomes reflected at the air-metal interface. In an attempt to improve the situation, one can try to reduce the reflection of the ferromagnetic layer using an anti-reflection (AR) layer. In fig. 5(d)–(f) the transmission, reflection and absorption of the same Fe/Co layer is shown using an additional layer on top of the ferromagnetic metal ($n_{AR} = 27$, $d_{AR} = 20\,\mu$m). Figure 5(e) clearly shows that using an AR-layer we can reduce the reflection to values below 5% outside of the resonance. This in fact leads to an enhancement of the transmission by about two orders of magnitude. As a possible realization of such an AR-layer could be the fine-tuning of $SrTiO_3$-based ceramics, which are known to show high values of the dielectric permittivity [29]. However, further improvement of the transmission using conventional ferromagnetic metals seems difficult. Another possible solution to this problem could be to use superconductors. In this case and in the millimeter range the conductivity will be purely imaginary, *i.e.* no internal absorption will be present. At the current stage of research this solution would also be difficult, because magnetism and (singlet) superconductivity tend to exclude each other.

Conclusions. – In conclusion, we have shown that the refractive index in natural ferromagnetic metals can be drawn negative close to the frequency of the ferromagnetic resonance. We have investigated pure cobalt and Fe/Co alloy both revealing negative values of the refractive index over a large temperature range and at different frequencies in the millimeter-wave range. Both metals show a large negative index of refraction at room temperature which is highly tunable by the external magnetic field.

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