Magnetotransverse Scattering of Surface Plasmon Polaritons

G. Düchs,[1] G. L. J. A. Rikken,[1] T. Grenet,[2] and P. Wyder[1]
[1]*Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung/CNRS, Boîte Postale 166, 38042 Grenoble Cedex 9, France*
[2]*Laboratoire d'Etudes des Propriétés Electroniques des Solides, CNRS, Boîte Postale 166, 38042 Grenoble Cedex 9, France*
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We show experimentally that the in-plane scattering of surface plasmon polaritons (SPP) is influenced by a perpendicular magnetic field. The average SPP flux is deflected into the direction perpendicular to both its initial propagation direction and the magnetic field direction. From a phenomenological point of view, this is an analogy to the Hall effect for electrons and a 2D equivalent of the photonic Hall effect.

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Surface plasmon polaritons (SPP) are p-polarized electromagnetic waves that propagate along a dielectric-metal interface [1]. They are carried by collective oscillations of the free carrier density in the metal, and their amplitudes decay exponentially with increasing distance away from the interface. In many aspects SPP can be regarded as two-dimensional electromagnetic waves. As their dispersion relation is very sensitive to the optical properties near the interface, SPPs have been used in surface sensors with monolayer sensitivity [2]. Recently, both theoretical and experimental interest in the propagation and scattering of SPP has rekindled. Many phenomena have been studied, such as SPP interaction with small defects [3,4], photonic band gaps for surface modes [5], SPP localization [6], SPP induced tunneling, etc. [7]. These studies may find application in novel photonic devices [8] and open new technological areas such as surface chemical analysis on the nanometer scale, two-dimensional optics of SPP, novel integrated SPP sensors, etc. Thus far, an influence of magnetic fields on the scattering of SPP has never been observed, although calculations suggest that such an influence ought to exist [9,10]. Our studies with three-dimensional light scattering in magnetic fields clearly established a deviation of a diffusive photon flux into the direction perpendicular to both the initial propagation direction of the photons and the external magnetic field [11,12]. This effect was baptized photonic Hall effect in view of its analogy to the electronic Hall effect. The question naturally comes to mind whether, in spite of differences in dimensionality and polarization state, a similar phenomenon could occur for the scattering of SPP in a magnetic field. In this Letter, we show experimentally that this is indeed the case and report the first observation of magnetotransverse scattering of SPP.

On a perfect flat interface, SPP propagate ballistically over macroscopic distances until they are absorbed in the metal. Interface roughness or inhomogeneities in the optical properties near the interface are perturbations acting on the SPP and lead to elastic scattering both in the plane of the interface and out of this plane. Consider the situation of Fig. 1: SPP are diffusively propagating with the initial direction from the top to the bottom. Fick's diffusion law relates the SPP flux density $\mathbf{J}$ to the local SPP density gradient $\nabla\rho$ according to $\mathbf{J} = D \cdot \nabla\rho$, where D is the diffusion tensor. For randomly shaped and randomly distributed scatterers, the in-plane scattering of the SPP is symmetric with respect to their initial direction of propagation and the SPP flux into two directions α and $-\alpha$ is equal: $\Delta\mathbf{J}(\alpha) \equiv \mathbf{J}(\alpha) - \mathbf{J}(-\alpha) = 0$. However, applying a magnetic field $\mathbf{B}$ perpendicular to the interface breaks the spatial symmetry in the interface plane. A net magnetotransverse SPP flux $\Delta\mathbf{J}(\alpha, B) \propto V\mathbf{B} \times \nabla\rho$ in this plane becomes symmetry allowed. In this equation, V is the Verdet constant of the scatterers; this is in essence the first-order coupling parameter between electromagnetic

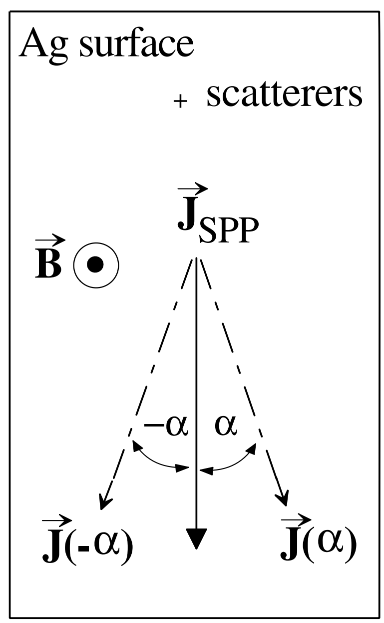

FIG. 1. SPP propagate on a silver surface with the initial direction $\mathbf{J}$ from top to bottom and scatter at surface irregularities. A perpendicularly applied magnetic field breaks the left-right symmetry and an additional net magnetotransverse flux $\Delta\mathbf{J}(\alpha, \mathbf{B}) \equiv \mathbf{J}(\alpha, \mathbf{B}) - \mathbf{J}(-\alpha, \mathbf{B}) \neq 0$ of scattered SPP becomes allowed.

waves and static magnetic fields and describes their magneto-optical activity [13]. Although there is no rigorous theoretical prediction for the magnitude of such an effect, dimensionality arguments suggest that the ratio of the magnetotransverse flux and the incident flux should be of the order of $GVBl$, where l is some characteristic length of the scattering process and G is a dimensionless geometry parameter. In the case of the three-dimensional photonic Hall effect, such a relation has indeed been observed with $G = 5 \times 10^{-3}$ [14]. The microscopic mechanism underlying this effect is the influence of the Lorentz force at the SPP oscillation frequency on the collective free electron motion, and thereby on the propagation and scattering of SPP. Note that the above symmetry consideration holds for diffusive SPP transport, whereas our experiment mostly deals with single scattering of SPP. As we did find a magnetotransverse SPP flux, this suggests that the difference between these two cases might not be decisive with respect to the symmetry breaking. This would be comparable to the three-dimensional photonic Hall effect, where a correspondence between the magnetotransverse scattering of one single Mie scatterer and diffusive transport, has been found [15].

In our experiment, SPP are excited by means of a diode laser ($\lambda = 633$ nm), on the surface of a thin silver film (thickness $d_{\mathrm{Ag}} = 60$ nm) on the bottom side of a glass prism [Kretschmann-Raether configuration, Fig. 2(a)]. The SPP propagate on the silver surface exposed to the air and are scattered by surface irregularities before retransforming into light. As they scatter elastically in all directions in the plane of the silver surface, light is reemitted not only in one direction but on the surface of a cone. This leads to the appearance of a bright circle when looking perpendicularly onto the metal film [Fig. 2(b)]. In this circle, the point at the very bottom corresponds to SPP that have maintained their initial direction of propagation in the metal film. The right part of the circle corresponds to SPP that have scattered more to the right, and the left part to SPP that have scattered more to the left inside the metal film. Because of the symmetry of the scattering in the silver film, the intensity profile of this bright circle must on average be left-right symmetric.

However, when we apply a magnetic field perpendicular to the plane of SPP propagation, this symmetry is broken. To observe this symmetry breaking experimentally, we compare the light intensities at two symmetrically situated positions at α and $-\alpha$ on the scattering intensity profile and measure their difference, $\Delta I(\alpha, B) \equiv I(\alpha, B) - I(-\alpha, B)$. The light at these two positions is collected and guided to photomultipliers by means of two optical fibers (diameter: 0.5 mm; numerical aperture: 0.5). The normalized value $\eta \equiv \Delta I/[I(\alpha) + I(-\alpha)]$ is a measure of the relative magnitude of the magnetotransverse SPP flux. In order to reliably measure this difference signal and to suppress artifacts, we apply an oscillating magnetic field $B(t) = B \cos \Omega t$ ($\Omega = 1.2$ Hz; $B \leq 0.5$ T) and perform phase-sensitive detection on ΔI (cf. Fig. 3) which yields in essence $\Delta I(\alpha, B) - \Delta I(\alpha, -B)$. To facilitate changing samples, the silver films were evaporated onto thin glass slides which were then optically contacted to a glass prism (BK7) by means of an index-matching liquid.

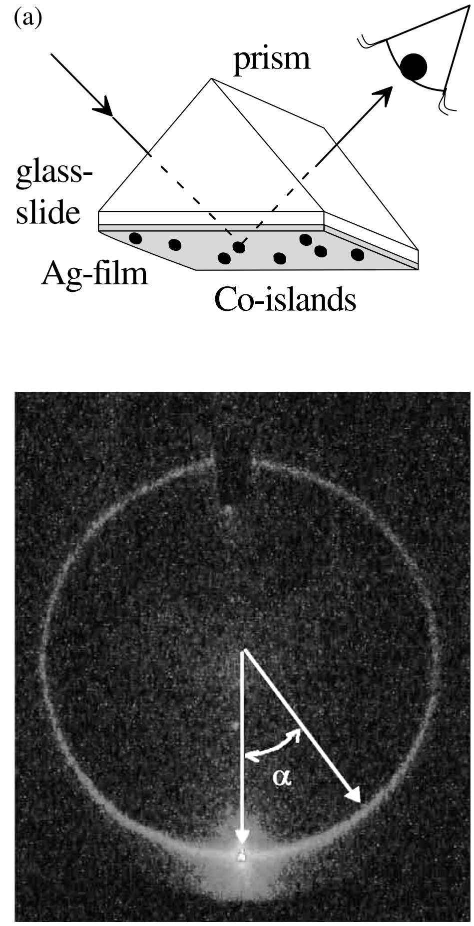

FIG. 2. The sample consists of a silver film with cobalt islands on it (a). It is supported by a glass slide that is optically connected to a prism. SPP are excited at the silver/air interface and scatter in all directions in that plane before retransforming into photons. Light is emitted on the surface of a cone and a bright circle can be seen (b). Without magnetic field, the intensity profile of this circle must be left-right symmetric. (The picture was taken with a glass half sphere instead of a prism.)

When the intrinsic roughness of the silver-air interface was the only source of SPP scattering, no significant values for a magnetically induced transverse SPP flux were observed: $\eta_{\mathrm{Ag}}/B \leq 10^{-5}\ \mathrm{T}^{-1}$. As the scatterers for this case should be regarded as either silver islands on a homogeneous silver film or air-filled depressions in the silver film, the Verdet constant of silver [16] or air [17], respectively, should determine the strength of the magnetotransverse scattering. As both are quite small, the absence of a significant magnetotransverse SPP scattering is not surprising. We therefore introduced additional scatterers with a high Verdet constant on the silver surface. Cobalt was chosen as a scatterer material because of its high

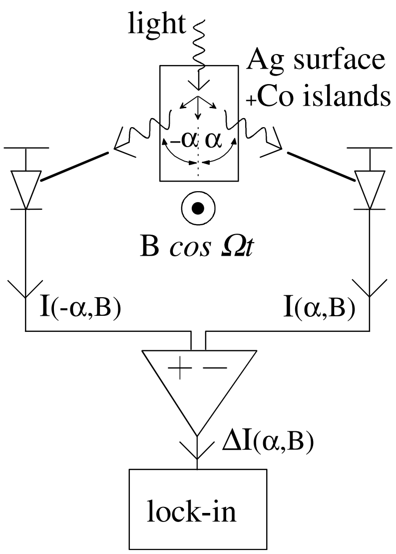

FIG. 3. Schematic setup of the experiment. SPP propagate on a silver surface which is exposed to an oscillating magnetic field. By cobalt islands, they are scattered in all directions in the plane. The reemitted light is captured at two opposite positions α and $-\alpha$ and the magnetotransverse SPP flux $\Delta I(\alpha, B) \equiv I(\alpha, B) - I(-\alpha, B)$ is detected with a phase-sensitive detection technique.

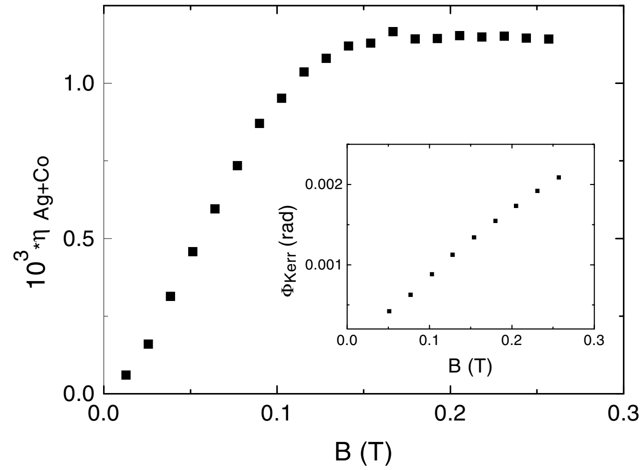

FIG. 4. The normalized magnetotransverse SPP flux $\eta_{\mathrm{Ag+Co}}$ as a function of the magnetic field for $\alpha \approx 11°$. For small magnetic fields, the effect is proportional to B. The saturation for magnetic fields larger than $B \simeq 0.15$ T is much more pronounced than the saturation of the polar Kerr rotation on the homogeneous cobalt film which is shown in the inset. The observed Kerr rotation at $B = 0.2$ T corresponds to a Verdet constant of 3.3×10^5 rad/m.

magneto-optical activity [18], and because of its ferromagnetic properties. This latter aspect implies that small external magnetic fields can evoke a large magneto-optical response. Cobalt deposition was performed by electron beam evaporation through a mask containing small circular holes. The resulting cobalt islands had a thickness of about $d_{\mathrm{Co}} = 70$ nm and a radius of $a = 3.0 \pm 1.0$ μm, as measured by scanning electron microscopy. The islands are randomly distributed with a density of about $3000/\mathrm{mm}^2$ corresponding to a mean distance between the islands of $l_{\mathrm{dist}} \approx 10$ μm. According to [1], the absorption length l_{abs} of SPP in a 60 nm silver film is of the order of 20 μm. This means that we mostly deal with single Mie scattering of the SPP by the cobalt islands. This has also been confirmed by a direct measurement of the propagation length l_{prop} of the SPP which shows that $l_{\mathrm{prop}} \simeq 10$ μm.

The magneto-optical properties of the cobalt were verified by polar Kerr rotation measurements on homogeneous cobalt films of the same thickness as the cobalt islands of the samples. (The polar Kerr effect involves the same polarizations as relevant to the SPP propagation and scattering.) As the cobalt films are polycrystalline with arbitrary crystallite orientations, our results are automatically averaged over the magnetic crystal anisotropy of cobalt. The result of such a measurement is shown in the inset of Fig. 4; the polar Kerr rotation, and thereby the Verdet constant, starts to saturate at the highest fields applied in this experiment. Note that, in this geometry, the demagnetization factors are close to unity and not exactly the same for the islands and the homogeneous film, so some scaling of the magnetic field axis may be necessary to translate the film results to island values. The observed Kerr rotation at $B = 0.2$ T corresponds to a Verdet constant of 3.3×10^5 rad/m, which is about a factor of 2 smaller than the literature saturation value on bulk cobalt [18].

The magnetotransverse SPP scattering by the cobalt-silver films was measured in the way as indicated above. In Fig. 4, $\eta_{\mathrm{Ag+Co}}$ is plotted as a function of the externally applied magnetic field for $\alpha \approx 11°$ and for scattering parameters as indicated above. Similar results have been obtained for other samples with differently shaped and distributed cobalt particles. From these measurements it is evident that a magnetotransverse flux of SPP exists for the case of cobalt scatterers. For small magnetic fields, the effect is proportional to B. Furthermore, for magnetic fields larger than $B \simeq 0.15$ T, we find a very clear saturation of the effect, much more pronounced than the saturation of the Kerr rotation on the homogenous cobalt film. This difference may be due to the difference in demagnetization factors for films and islands. In addition, the very large value of $V_{\mathrm{Co}}a$ realized in our experiment may also imply that we have entered a regime where the magnetically induced changes of the optical properties of the system can no longer be regarded as small perturbations. In this case, η need no longer be linearly dependent on $V_{\mathrm{Co}}B$. In the measurement presented in Fig. 4, η reaches a value of 2.5×10^{-3} in the regime of saturation. Note that this saturation value is dependent on the exact parameters of the Ag surface and the Co islands. We point out that all measured η values represent an underestimate of the magnetotransverse SPP flux due to the cobalt islands, as part of the intensity measured at positions α and $-\alpha$ is due to SPP scattering by inhomogeneities in the silver

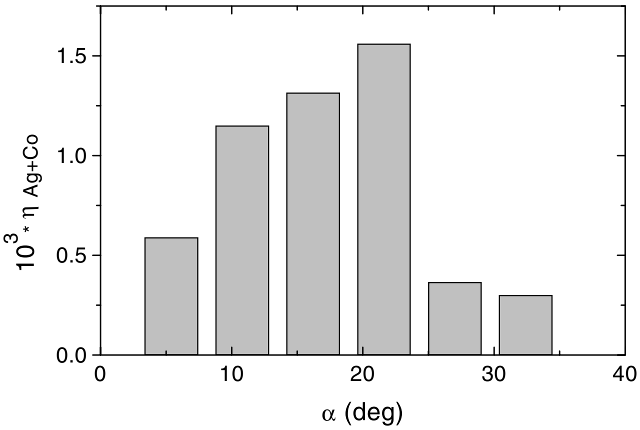

FIG. 5. The normalized magnetotransverse SPP flux η_{Ag+Co} in the regime of saturation as a function of the scattering angle α. A maximum is observed for $\alpha \approx 20°$. Note that there is a systematic uncertainty of 15% on α which might change the scaling of the axis.

film itself, that shows no magnetic field dependence. The order of magnitude of η roughly corresponds to the values obtained for the 3D photonic Hall effect with saturated ferromagnetic scatterers where also values as large as $\eta \approx 10^{-3}$ have been experimentally observed, albeit in the regime of multiple Rayleigh scattering [19]. This suggests that magnetotransverse scattering of electromagnetic waves is mostly independent on details of the scattering geometry.

Figure 5 shows, for the same sample as in Fig. 4, the dependence of the normalized magnetotransverse SPP flux on α, i.e., the differential magnetoscattering crosssection. We observe a maximum at an angle of about $\alpha_{\max} \approx 20°$. The differential scattering cross section for single scattering of SPP by a small circular surface defect was calculated in Ref. [3] but no magnetic field had been taken into account; and as the cobalt islands of our samples are large compared to the wavelength, this calculation does not apply to our experiment. However, in a scattering cross-section model for the three-dimensional photonic Hall effect [14,15], the influence of the perpendicular magnetic field on photon scattering could be described by rotating the differential scattering cross section around the magnetic field axis over an angle $\beta \approx VBs$, where V is the Verdet constant of the scattering medium, and s is a characteristic scattering length. This results in a net magnetotransverse current of photons, i.e., a photonic Hall effect. If we assume that in the case of SPP scattering the magnetic field will affect the differential scattering cross section in a similar way, this would also lead to a net magnetotransverse flux of SPP. In order to evaluate this description for SPP scattering by our cobalt islands, a calculation of the differential scattering cross section for this case, including the size dispersion, is needed.

In conclusion, our experiments have shown the existence of magnetotransverse scattering of surface plasmon polaritons for the case of single Mie scattering from cobalt islands deposited on a silver surface. We find reasonable agreement with simple estimates for such an effect. Related SPP magnetoscattering phenomena, such as Rayleigh scattering, multiple scattering, or coherent backscattering of SPP in magnetic fields, now could be studied. The diffusion experiments would be particularly interesting as, in general, diffusion in two dimensions is known to be drastically different from diffusion in three dimensions [20].

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