Brief Communication Arising to:
Experimental demonstration of the microscopic origin of circular dichroism in two-dimensional metamaterials, Nat.Commun. 7 (2016).

Circular dichroism and two-dimensional metamaterials

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The relation between geometrical concept of chirality, the property of objects that cannot be superposed to their mirror images, and the handedness of polarized light waves has been fascinating researchers since 1848 [1], when Pasteur realized that handpicked oppositely shaped crystals of tartaric acid had opposite values of optical activity. In the last years the interest in studying the interaction of light with chiral objects is stronger than ever, since modern fabrication techniques permit to engineering nano objects with on demand geometrical features.

In a recent publication in Nature Communications [2] it is reported that circular dichroism can emerge in a two-dimensional (2D) metasurface. Authors claim that the presence of circular dichroism in such structure is proved by the transmission difference between left and right circular polarized light. This is incorrect.

Circular dichroism is the difference in the absorption of left-handed circularly polarised light (L-CPL) and right-handed circularly polarised light (R-CPL) but, in general, CD cannot be measured by illuminating alternatively a material with L-CPL and R-CPL and analyzing the differential response. This simple approach seems intuitive but, in general, it is incorrect because it is not in agreement with the definition of CD.

The propagation of light in a material is a continuous process, and cannot be presented as a "one-shot" interaction. As light propagates in the medium, the polarization of the wave progressively changes due to effects such as linear retardation, linear diattenuation or depolarization. As a result, a beam of light, initially circularly polarized, changes its polarization as it propagates and, and when it is absorbed it is not circularly polarized anymore. Only in an isotropic chiral medium the circular polarization is an eigenmode of propagation, which means that the emerging wave is still circularly polarized and it is absorbed as a circularly polarized wave at any point of the optical path. This is the single case where CD can be measured directly from the differential transmittance of L-CPL and R-CPL. In all other situations it is advisable to go for polarimetric solutions based on the determination of the complete Mueller matrix [3]. If such full polarimetric methods are not available, errors in the assessment of CD can be often identified by repeating optical measurements with the sample turned over: the same CD should be measured regardless from which side the metasurface is examined.

True CD signals in *apparently 2D* metasurfaces are possible but they always arise due some hidden 3D character that arises, for example, from the dissymmetric effect of the substrate or from the smoothing of the edges of the real metasurface [4]. An ideal 2D metasurface is incompatible with CD. 2D

metasurfaces without four-fold rotational symmetry can show asymmetric transmission of CPL, an effect different from CD [5] that arises from the coherent superposition of misaligned refraction and dissipation modes of different parts of the nanostructure. This can be the case of the samples investigated in [2].

[1] L. Pasteur, "Mémoire sur la relation qui peut exister entre la forme cristalline et la composition chimique, et sur la cause de la polarisation rotatoire (Memoir on the relationship which can exist between crystalline form and chemical composition, and on the cause of rotary polarization)," *Comptes rendus de l'Académie des sciences* (Paris), 26, 535–538 (1848).

[2] A. B. Khanikaev, N. Arju, Z. Fan, D. Purtseladze, F. Lu, J. Lee, P. Sarriugarte, M. Schnell, R. Hillenbrand, M. A. Belkin, G. Shvets. "Experimental demonstration of the microscopic origin of circular dichroism in two-dimensional metamaterials," Nat. Commun. 7 (2016).

[3] O. Arteaga, "Mueller matrix polarimetry of anisotropic chiral media", PhD Thesis, Univ. Barcelona (2010).

[4] O. Arteaga, J. Sancho-Parramon, S. Nichols, B. M. Maoz, A. Canillas, S. Bosch, G. Markovich, and B. Kahr, "Relation between 2D/3D chirality and the appearance of chiroptical effects in real nanostructures," Opt. Express 24, 2242-2252 (2016)

[5] Note that despite this effect is different from CD it is confused with CD if one tries to assess CD only by the measuring the differential transmittance of L-CPL and R-CPL.