Disorder-induced material-insensitive optical response in plasmonic nanostructures: vibrant structural colours from noble metals

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Materials show various responses to incident light, owing to their unique dielectric functions. A well-known example is the distinct colours displayed by metals, providing probably the simplest method to identify gold, silver and bronze since ancient times. With the advancement of nanotechnology, optical structures with feature sizes smaller than the optical wavelength are routinely achieved. In this regime, the optical response is also determined by the geometry of the nanostructures, inspiring flourishing progress in plasmonics, photonic crystals and metamaterials. Nevertheless, the nature of the materials still plays a decisive role in light-matter interactions, and this material-dependent optical response is widely accepted as a norm in nanophotonics. Here, we propose and realise a counterintuitive system - plasmonic nanostructures composed of different materials but exhibiting almost identical reflection. The geometric disorder embedded in the system overwhelms the contribution of the material properties to the electrodynamics. Both numerical simulations and experimental results provide concrete evidence of the insensitivity of the optical response to different plasmonic materials. The same optical response is preserved with various materials, providing great flexibility of freedom in material selection. As a result, the proposed configuration may shed light on novel applications ranging from Raman spectroscopy, photocatalysis to nonlinear optics.

1 Introduction

The dielectric function (or the refractive index) is a dimensionless parameter quantitatively describing the interaction between light and a certain material. Differences in atomic or molecular structures induce different displacement fields under an applied electric field, producing unique dielectric functions of

materials [1]. Consequently, the variation in the spatial distribution of dielectric function results in different solutions of Maxwell equations, i.e., material-dependent optical responses. This results in distinct intrinsic colours for different metals, providing the simplest way for the identification with the naked eye. Owing to the rapid blossoming of nanofabrication and nanotechnology, structures with feature sizes comparable to or smaller than the optical wavelength can be routinely achieved. In contrast to their bulk counterparts, the nanoscale geometry significantly impacts the optical properties of a material. Gold nanostructures do not necessarily generate golden colour, but produce rainbow-liked colours[2, 3, 4] and even black ones [5, 6]. Various nanophotonic systems including plasmonic structures, photonic crystals and metamaterials [7, 8, 9] have been realised with unprecedented optical properties significantly different from the intrinsic features of bulk materials. However, even under this regime, the type of materials (or the value of refractive index) still plays a substantial role in the optical response of micro/nano structures [10, 11, 12]. For example, the quality factor of the resonance of metal nanostructures is mainly determined by the complex dielectric function under the quasi-static approximation [10]. As a result, a different design is required for switching from one material platform to another, even for achieving similar functionality in photonics[13, 14].

In this article, we propose and realise a highly counter-intuitive platform in nanophotonics, a disordered network composed of different plasmonic materials but with almost identical optical response. By virtue of random scattering, disordered structures are insensitive to optical parameters such as wavelength, incident angle and polarisation [15, 16, 17, 18, 6]. With judicious designs, disordered networks facilitated a repertoire of applications, including broadband reflector [19], structural colours [20, 21, 22, 23, 24], photocatalysis [25], optical filters [26] and lasers[27, 28, 29]. Here, we extend the insensitivity to the material's property in optics, demonstrating that disorder can effectively mitigate the material dependence of the optical response. When combined with judiciously engineered optical environment[30], disordered plasmonic nanostructures composed of a variety of materials produce nearly identical refection spectra. Equivalently, indistinguishable colours are generated from nanostructures composed of different metals. Based on *ab initio* simulations, we prove that similar optical responses can be obtained from a diverse material library including silver (Ag), gold (Au), aluminium (Al), chromium (Cr), copper (Cu), nickel(Ni), palladium(Pd), platinum(Pt), Ruthenium (Ru) and titanium nitride (TiN). Further experimental verification is implemented with four commonly-used metals (Au, Ag, Pd and Pt). Considering the strong light-matter interaction in the plasmonic nanostructures and unique chemical / electrical / mechanical properties of different materials, the proposed platforms may inspire novel designs for a repertoire of applications ranging from surface-enhanced Raman spectroscopy[31, 32], harmonic generation[33] to photocatalysis[34, 35].

2 Results

With the conventional wisdom, it is believed that the material property plays an indispensable role in the optical response. **Figure 1** provides an example of material-dependent absorption cross-sections Q_{abs} of metallic nanoparticles. Here, four different plasmonic materials, namely Ag, Au, Pt and Pd are selected. Figure 1a-d show the complex refractive indices $\tilde{n} = n + ik$ in the visible region, respectively. Figure 1e-h show the corresponding absorption cross-sections calculated from FDTD simulations, with the diameter of the spherical nanoparticle fixed to 10 nm. The difference in atomic structure results in the variation of the refractive index for different materials, directly leading to distinct optical responses in absorption. The line shapes of the cross-sections of Pd and Pt share some similarity, owing to the comparatively small difference in in their refractive indices $\tilde{n}$. Meanwhile, the Ag and Au nanoparticles have completely different cross-sections, as a result of the large discrepancy in $\tilde{n}$. Consequently, the type of material directly determines the optical dynamics, which matches our physical intuition. Similar effect is observed for scattering cross-sections, with more details can be found in Supplementary Note 1.

However, the material-dependent behaviour can be counterintuitively eliminated, with the aid of disorder in the geometry. Previously, we developed a mechanism that can manipulate reflection spectra with desired reflection peaks through disordered plasmonic nanostructures[30]. Here, we utilise disor-

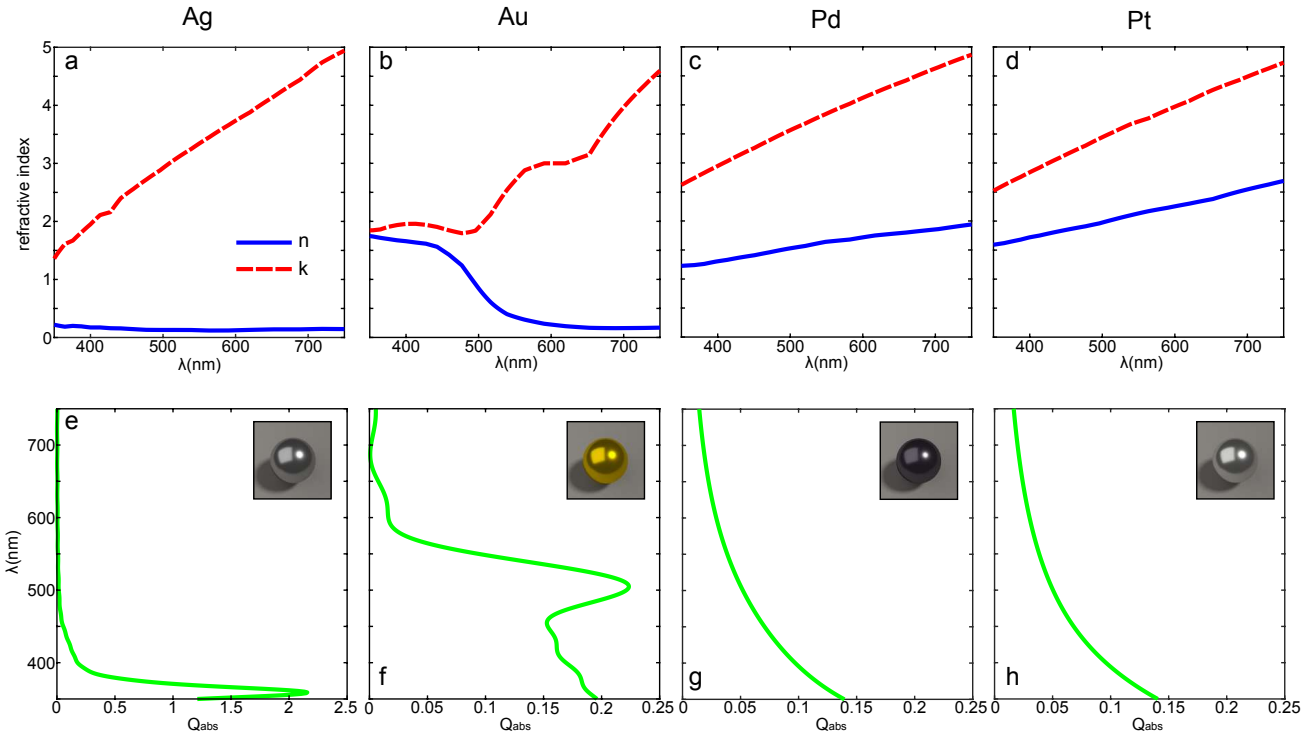

Figure 1: Material-dependent optical response. **a-d.** Complex refractive indices $\tilde{n} = n + ik$ for different materials a) Ag, b) Au, c) Pd and d)Pt. **e-h** Absorption cross-section (Q_{abs}) calculated based on FDTD simulations for e) Ag, f) Au, g) Pd and h) Pt. The size of the spherical nanoparticle is 10nm.

dered plasmonic nanostructures on a metallic mirror [30] to investigate the optical response with different materials. **Figure 2**a illustrates the schematic of the system. Disordered structures are deposited on a transparent dielectric layer, with a metallic substrate underneath. The size, shape and positions of the nanoparticles are randomised, endowing disorder into the system. The dielectric layer with fixed thickness t forms a Fabry-Perot resonator. The disorder enables the plasmonic structures on top to be an effective broadband absorber. All photons are absorbed by the plasmonic structure, except the ones trapped in the Fabry-Perot resonator. The dielectric layer selectively protects the photons with frequencies around the resonance and release them as reflection. As a result, desired reflection spectra are achieved with reflection peaks located at resonances of the Fabry-Perot cavity.

Here we focus on the effect of material composition of the plasmonic structures, selecting different metals for the disordered structures. In Figure 2, the four materials Ag, Au, Pd and Pt are investigated numerically. Details of the simulations can be found in the Supplementary Note 2. Figure 2b-d present the reflection spectra with different dielectric thicknesses. The black dashed line is the averaged value of all the spectra, working as a guide for comparison. Intriguingly, the reflection spectrum remains almost the same for disordered nanostructures composed of different materials, in sharp contrast with the optical response shown in Figure 1e-h. Despite the apparent dissimilarity in refractive indices among Ag, Au and Pd (Pt) shown in Figure 1a-d, the line shape of the reflection remains nearly identical with only minute differences. The disorder in geometry drastically mitigates the contribution of the refractive index to the optical dynamics of the system. Such a disorder-induced material-independent response is valid for all three thickness shown in Figures. 2b-d. In Figure 2e, the corresponding colours are calculated from the reflection spectra, with colours from two additional thicknesses provided. Due to the high sensitivity to colours, the difference in colour is more obvious to the naked eye. The negligible difference in the spectrum between Au and Pd turns into a subtly discernible variation in colour. A similar material-independent effect is unambiguously illustrated with other metals including Al, Cr, Co, Cu, Ni and even a compound (TiN). More details can be found in Supplementary Note 3. Simulations with different sets of random variables are demonstrated in Supplementary Note 4, confirming that the insensitivity is induced by disorder other than a specific setup.

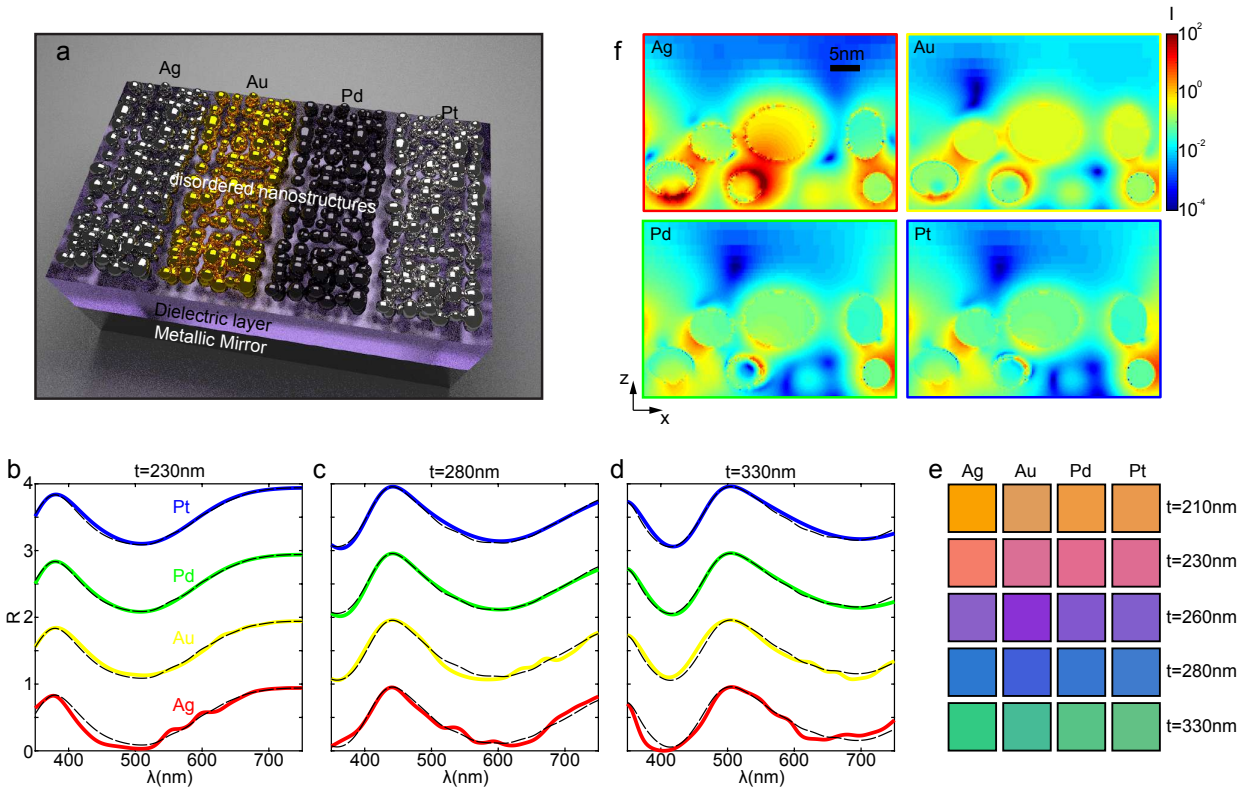

Figure 2: Numerical simulations of the material-independent optical response. **a** Schematic of the proposed structure - disordered plasmonic nanostructures on a metallic mirror with a transparent dielectric with thickness t between them. **b-d** Reflection spectra of the samples with different thickness b) t=230nm; c) t=280nm; d) t=330nm. Four different materials are showed; Ag, Au, Pd and Pt. Black dashed lines are guides for comparison. **e** The colour calculated from the reflection spectra from b-d). Two additional thickness are included. **f** Spatial distribution of light intensity I near the nanoparticles.

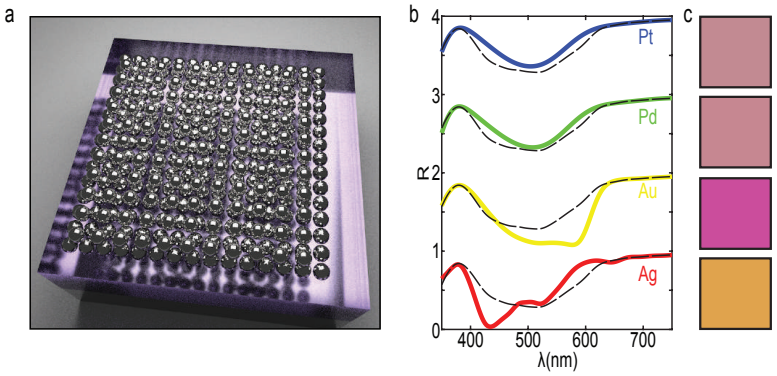

Figure 3: Comparison with ordered nanostructures with material-dependent optical response. **a** Schematic of a sample composed of plasmoinc clusters without disorder. **b** Corresponding reflection spectra based on four different materials; Ag, Au, Pd and Pt. The thickness of the dielectric layer is fixed to 330nm. **c** Corresponding colours calculated from spectra in b).

The collective behaviour from disordered structures structures underlies the material-independent response. The dynamics for individual nanoparticles still varies. The effect is demonstrated in Figure 2f, showing the spatial distribution of the light intensity I around the nanoparticles. The thickness t is 330 nm and the wavelength of the incident light is 505 nm. In the near field, the spatial distribution varies dramatically for different materials. However, the reflection contributed by random scattering from all nanoparticles in the farfield shares a high level of similarity, as illustrated in Figure 2d.
To clarify the dominant role played by disorder in reducing the material dependence, we implement further numerical analysis, which is summarised in **Figure 3**. A configuration similar to Figure2a but with-

out disorder is investigated, as shown in Figure 3a. All nanoparticles have a spherical shape with a fixed size, arranged in a periodic array. The thickness of the dielectric layer is selected as 330 nm. Figure 3b shows the reflection spectra of the ordered system composed of different materials, while Figure 3c illustrates the corresponding colours. Without disorder, the choice of material contributes significantly to the reflection spectra. The structures composed of Pd and Pt share similar reflection, due to the small difference in refractive index (Figure 1c-d). However, the spectra of Ag and Au are dramatically different, producing reddish and yellowish colours respectively.

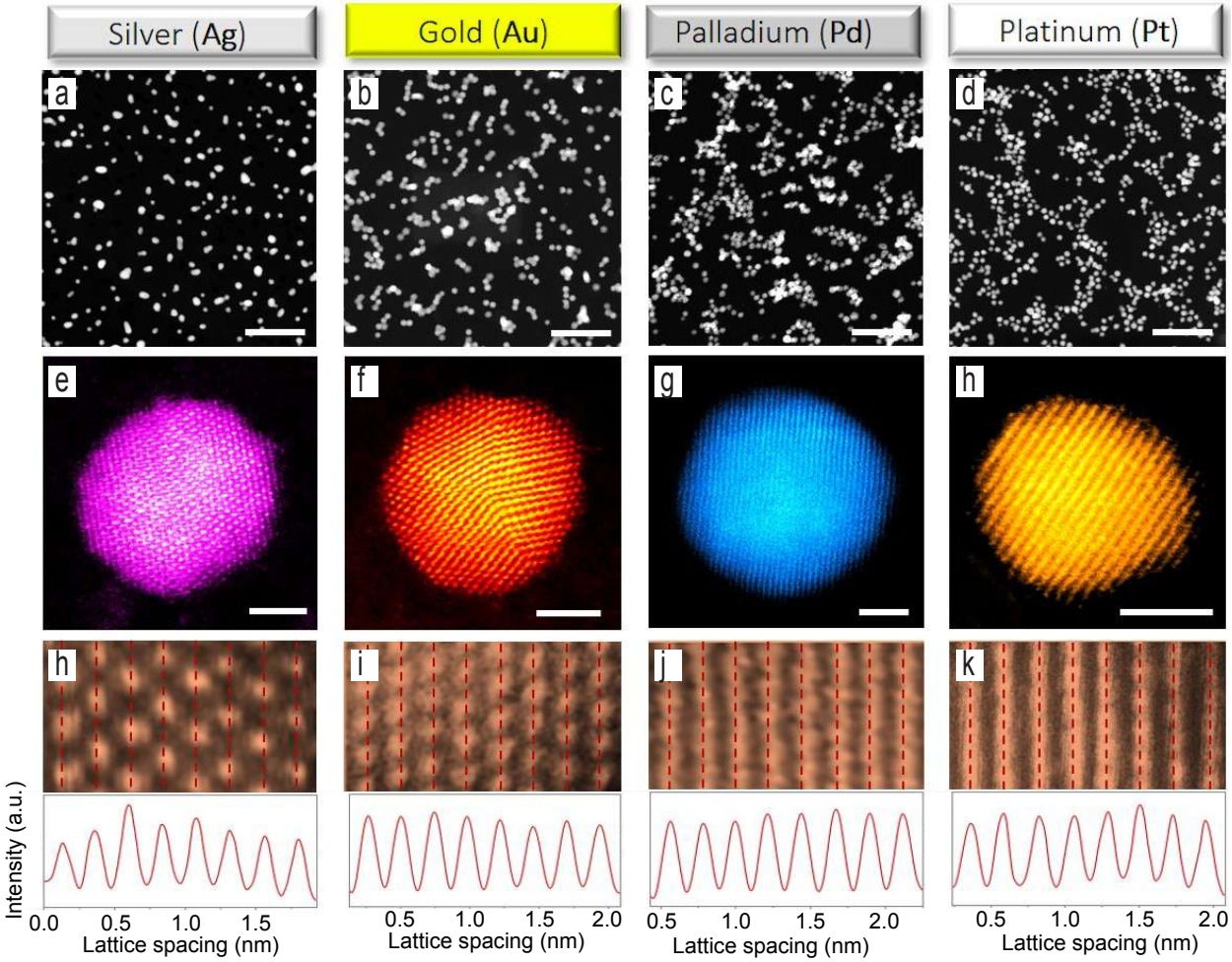

Figure 4: **a-d** Typical STEM images of different metallic nanoclusters; a) Ag, b) Au, c) Pd and d) Pt. Scale bar: 100 nm. **e-h** Corresponding high magnification STEM of individual metallic nanocluster. Scale bar: 2 nm. **i-l** Upper panels: Zoomed in STEM images of atomic columns within individual Ag, Au, Pd and Pt nanocluster. Lower panels:The integrated pixel intensities along (111) directions (which is perpendicular to the planes). The peaks and valleys represent the atoms and gaps, respectively. The spacing of (111) planes is averaged over 8 atomic layers for high accuracy.

For the experimental realisation, we utilise gas-phase cluster beam deposition to form the disordered nanostructures [36, 37]. The disorder in position and shape of nanoclusters is a intrinsic feature of the fabrication process. Moreover, the cluster beam technique offers a feasible way to produce clusters composed of different materials. More details are provided in the Experimental section. In the following experiments, four different plasmonic materials are investigated; Ag, Au, Pd and Pt. **Figure 4** provides a comprehensive characterisation of the clusters formed by different materials. The metallic nanoclusters (Ag, Au, Pd and Pt) generated from cluster beam deposition system are characterised by scanning transmission electron microscopy (STEM). Figures 4a-d show STEM images of the disordered metallic nanocluster systems, demonstrating the disorder embedded in both the shape and position of the nanoclusters. As a further illustration of the disorder in position, we perform a two-dimensional Fast Fourier Transform (2D-FFT) based on the positions of the nanoclusters, as shown in Supplementary Note 4.

Nanoclusters with different compositions are prepared with basically the same size distribution. The averaged sizes of the Ag, Au, Pd and Pt nanoclusters are 11.09 nm, 11.51 nm, 11.32 nm and 10.92 nm, respectively. More details can be found in Supplementary Note 4. Figures 4e-h show typical high magnification HAADF-STEM images of single Ag, Au, Pd and Pt nanoclusters, respectively. Figure 4 i-l investigate the (111) interplanar spacing for Ag, Au, Pd and Pt nanoparticles respectively. Central regions are selected from the STEM images (upper panels) to avoid possible surface defects as well as blurry boundaries between metal and the substrates. The lower panels show the integrated pixel intensities correspondingly. The averaged pristine Ag, Au, Pd and Pt (111) interplanar spacings are 2.36Å, 2.41Å, 2.26Å and 2.29Å, matching previously-reported interplanar constants [38].

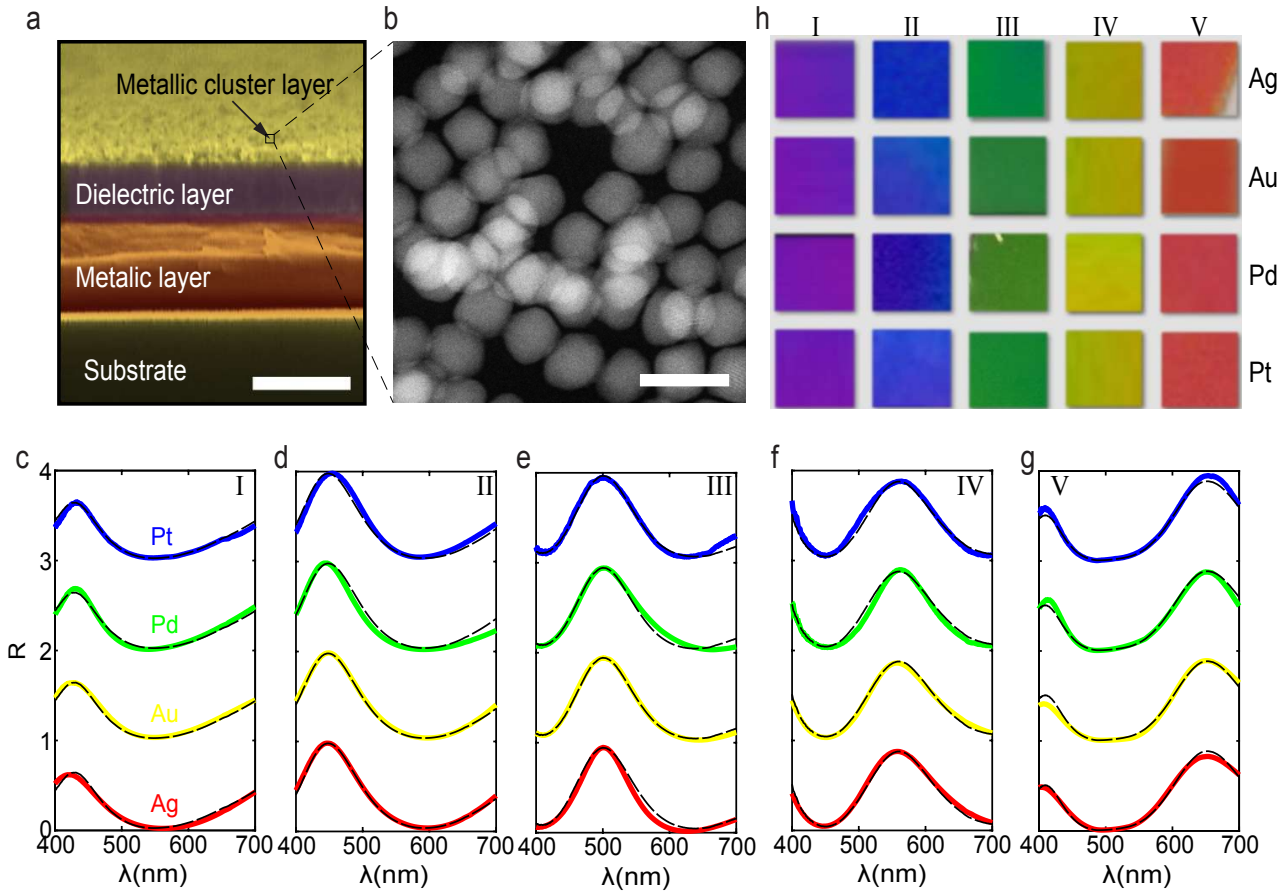

Figure 5: Experimental realisation of material-independent optical response. **a** SEM image of the cross-section of a typical sample consisting of a metallic nanocluster layer separated from a silver film with a LiF spacer layer. Scale bar: 100 nm. **b** Typical high magnification STEM image of Pd nanoclusters. Scale bar: 20nm. **c-g** Reflection spectra of the samples with five different spacer thicknesses. Nearly identical reflection is observed for the different materials; Ag, Au, Pd and Pt. Black dashed lines are guides for comparison. **h** Photos of corresponding samples with different thicknesses and materials. The thickness of the spacer t: I) 290nm, II) 330nm, III) 370nm, VI) 410nm, V) 430nm.

Figure 5 summarises the experimental results of the study of the material-independent optical response. Figure 5a shows a typical Scanning Electron Microscope (SEM) image of the structure depicted in Figure 2a. Here, the metallic clusters are composed of Pd. Figure 5b shows a typical high magnification STEM image of Pd nanoparticles, further demonstrating the disorder embedded in both the shape and position. Figures 5c-g provide the reflection spectra from five samples (I to V) with different thicknesses of the dielectric layer. Interestingly, the variation among the samples composed of different materials is even smaller than that shown in numerical simulations. We attribute the enhanced independence to the stronger disorder in experiments (than in simulations), associated the complex shape of nanoparticles. Similar to the numerical results shown in Figures. 3a-c and Figures. 2b-e, disorder can help to overwhelm the heterogeneous material properties. Consequently, almost identical colours are produced

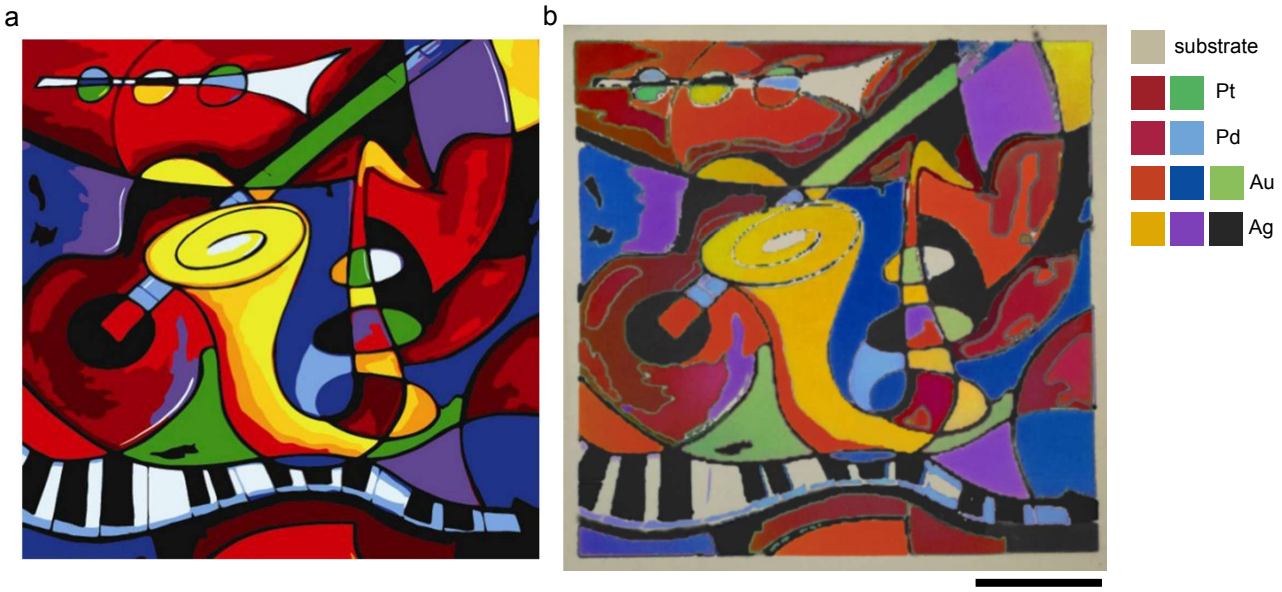

Figure 6: Colourful pattern developed by disordered multi-material nanostrucutres. **a** Original colourful pattern. **b** The fabricated structures incorporated with disordered cluster layers. Four different metallic nanoclusters are utilised; Ag, Au, Pd and Pt. Different colours are realised from different metals as shown in inset on the right. Scale bar: 10mm.

for structures with various materials, as demonstrated in Figure 5h. Based on these structural colours, it is impossible to identify the material of the plasmonic structure, even with a complete knowledge of the refractive index and the geometry. A material-independent optical response is thus realised with the aid of disorder.

We implement a further demonstration of the material-insensitive optical behaviour by using the plasmonic structures to replicate a colourful pattern. **Figure 6**a is the original pattern composed of several colours. The replicated image is formed from disordered nanostructures shown in Figure 6b. Four different metals are utilised to form the disordered structures; Ag, Au, Pd and Pt. Different colours are formed by different materials, as shown in the inset of Figure6b. Despite the different materials used (inset of Figure 6b), the colours are solely determined via variation in the thickness of the underlying Fabry-Perot spacer layer. The same optical function is realised by different material platforms without any performance degradation. In Supplementary Note 7, we provide another demonstration of pattern colouration, realising the same colour with different materials.

3 Conclusion

In this article, we propose and realise a highly counter-intuitive system - disordered nanostructures with a material-independent optical response. The structural colours generated show negligible variation by substitution of different materials. The geometric disorder dramatically reduces the dependence of the refractive index in optical dynamics. From numerical simulations, we demonstrate that the mechanism works for different plasmonic materials ranging from Au, Al, Cr, Cu, to Ni, Pd, Pt, Ru and even TiN. Meanwhile, experimental verification is illustrated with four of metals (Ag, Au, Pd and Pt), producing indiscernible structural colours.

For a practical device, the optical properties may not be the only concern. The material cost, stability, electrical and chemical properties also play substantial roles in the device performance. With a material-independent optical response, the optical design is isolated from the other considerations, offering potential for the improvement of other properties. For example, Au or TiN can be selected for stability, while Al can be chosen for cost. Cu, Cr rand Ru are ideal candidates for photocatalysis [34, 39, 40], when they share optical absorption similar to Au or Ag. Broadband absorption (black colour) is achieved for disordered networks composed of Ag, Au, Pd and Pt, with details given in Supplementary Note 8.

The material dependence in the far-field optical properties can also be reduced by minimising the light-matter interactions (e.g. similar reflection can be achieved for the same nanostructures on top of different substrates[22]). Here, we propose and realise a system achieving material insensitivity and strong light-matter interactions simultaneously. Given the versatile applications of disordered nanostructures [19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29], a disorder induced material-independent response may shed light on novel systems with a flexible material library.
In this article, our investigation of the optical response is limited to the linear regime. The nonlinear effect in plasmonic structures composed of different materials has been the subject of intensive research during the past decade[41, 42]. It will be interesting to investigate to which extent the disorder can lead to the similarity of second/third harmonic generation in the farfield from one material to another. Also, the current library only contains plasmonic materials. Further investigation of dielectric materials such as Si, GaN and TiO_2 will potentially extend the usefulness of the mechanism demonstrated to a variety of applications [43, 44]. Besides, it is also interesting from a perspective of mathematics, investigating the existence of a group of differential equations (including Maxwell's equations) providing insensitive solutions in the farfield, in spite of the variation of boundary conditions.

4 Experimental Section

Fabrication of metallic nanoclusters:
Silver (Ag) and lithium fluoride (LiF) films were deposited in a thermal evaporation system (SinoRaybo Nanoscience and Nanotech Co., Ltd). All evaporations were performed at a high vacuum, the working pressure of the system is about $1.5 \cdot 10^{-7}$ Torr. The deposition rate and film thickness were monitored by a quartz crystal microbalance, demonstrated in our previous work [30]. A gas-phase cluster beam deposition process was used to deposit metallic clusters (Ag, Au, Pd and Pt) on top of Ag film/spacer layer structure with details given in Supplementary Note 4. In our setup, the maximum size of the sample can be 15×15 cm^2, only limited by the size of the gas-phase cluster beam deposition equipment.

Sample characterisation:
The STEM investigation was performed using a JEOL instrument (JEM2100F) with a spherical-aberration corrector (CEOS GmbH). The images were acquired using high-angle annular dark field and bright field detectors. The thickness of the spacer was characterized by scanning electron microscopy (SEM, Hitachi S4800). Reflection spectra were collected over the UV-visible spectrum (350-800 nm) using a Zolix λ-300 spectrograph and monochromator (Zolix Instruments Co., Ltd.) in conjunction with a UV-VIS-NIR light source (Deuterium-Tungsten Halogen Source, Ocean Insight).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Contributions
These authors contributed equally: Peng Mao, Changxu Liu. C.L. and P.M. conceived the idea and designed the experiment. C.L. developed the model, performed the numerical simulations. P.M., Y.N., Y.Q., F.S., M. H. and R. E. P. performed the sample fabrication and characterisation. C.L. and P.M. wrote the manuscript with the input from S.A.M., S.Z. and R. E. P. . S.Z., S.A.M. and C.L. supervised the project. All authors discussed the results.

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