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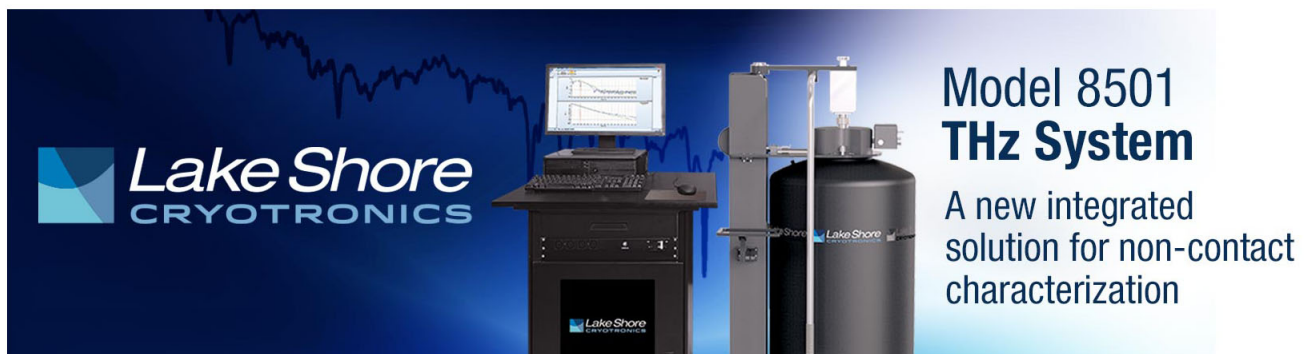
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## Excitons in a mirror: Formation of “optical bilayers” using MoS<sub>2</sub> monolayers on gold substrates

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We report coupling of excitons in monolayers of molybdenum disulphide to their mirror image in an underlying gold substrate. Excitons at the direct band gap are little affected by the substrate whereas strongly bound C-excitons associated with a van-Hove singularity change drastically. On quartz substrates only one C-exciton is visible (in the blue) but on gold substrates a strong red-shifted extra resonance in the green is seen. Exciton coupling to its image leads to formation of a “mirror biexciton” with enhanced binding energy. Estimates of this energy shift in an emitter-gold system match experiments well. The absorption spectrum of MoS<sub>2</sub> on gold thus resembles a bilayer of MoS<sub>2</sub> which has been created by optical coupling. Additional top-mirrors produce an “optical bulk.”

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The two-dimensional transition metal di-chalcogenide molybdenum disulfide (MoS<sub>2</sub>) has recently attracted much interest as a prominent example of a new class of monolayer materials with remarkable physical properties. Molybdenum covalently bonded to sulphur can form two-dimensional sheets that are linked to each other by weak van der Waals interactions.<sup>1,2</sup> This weak interlayer interaction makes MoS<sub>2</sub> highly suitable as a lubricant in industrial applications and provides the possibility to fabricate monolayers of sulphur-sandwiched molybdenum atoms by micromechanical cleavage, similar to the fabrication of graphene.<sup>3</sup> Unlike graphene, MoS<sub>2</sub> is in its bulk form a semiconductor with an indirect band gap at the  $\Gamma$ -point of the Brillouin zone. The electronic band structure of MoS<sub>2</sub> changes with thickness. Thinning the material down to a single monolayer transforms it into a direct gap semiconductor with the band gap located at the  $K$ -point. This electronic transition has been thoroughly studied both theoretically<sup>4–10</sup> as well as experimentally, for example, by absorption,<sup>11,12</sup> photoluminescence,<sup>11–13</sup> and Raman<sup>14–17</sup> spectroscopy. The optical band gap of a monolayer MoS<sub>2</sub> is in the visible part of the spectrum with an energy of 1.9 eV.<sup>11</sup> At the  $K$ -point, the valence band is split into two states due to the lack of inversion symmetry in the MoS<sub>2</sub> monolayers resulting in a strong spin-orbit interaction.<sup>7</sup> Transitions from both states to the conduction band have been observed as distinct  $K$ -excitons.<sup>8,11,18,19</sup> These special electronic and symmetry configurations of MoS<sub>2</sub> monolayers open the possibility for new spin valley physics.<sup>20,21</sup> Monolayers of MoS<sub>2</sub> are of interest for many technological applications ranging from logic circuit designs,<sup>22–25</sup> photodetectors,<sup>26,27</sup> batteries,<sup>28,29</sup> and catalytic applications<sup>30,31</sup> to solar cells.<sup>32,33</sup> For all these applications, it is highly desirable to engineer the electronic band structure

of MoS<sub>2</sub>. Tuning the electronic properties has been demonstrated electronically<sup>34</sup> and by using strain.<sup>35–37</sup>

In this paper, we show that the optical properties of MoS<sub>2</sub> can be strongly influenced when placed in close proximity to a gold-coated surface. We investigate the effect of metal surfaces on MoS<sub>2</sub> monolayers with dark field optical spectroscopy and compare results to MoS<sub>2</sub> monolayers on quartz. Whereas excitonic states near the  $K$ -point are little altered, drastic changes in the optical spectrum are observed associated with higher-energy bound transitions located between the  $K$ - and  $\Gamma$ -points of the Brillouin zone. We show that these arise not from doping effects, but from the dipole coupling of the excitons to their image charges.

In our studies, monolayers of MoS<sub>2</sub>, confirmed by Raman spectroscopy showing a frequency difference between the E<sub>2g</sub><sup>1</sup> and A<sub>1g</sub> mode of  $\Delta\omega = 19 \text{ cm}^{-1}$ ,<sup>15,38</sup> are prepared by chemical vapour deposition (CVD) on a sapphire substrate. The detailed fabrication process can be found elsewhere.<sup>39–42</sup> Monolayers are transferred to a quartz substrate or to a polished silicon substrate which is coated with 10 nm Cr and 100 nm Au by electron beam evaporation. A standard transfer method using poly(methyl methacrylate) (PMMA) is employed.<sup>40</sup> Additional samples are fabricated with an extra layer of 15 nm Au on top of the MoS<sub>2</sub> sheet, sufficiently thin to allow for spectroscopic measurements through this layer. The evaporation rate is chosen to be slow enough to ensure that a smooth continuous gold surface is formed, which is confirmed with scanning electron microscopy (SEM).

Scattering of light from the MoS<sub>2</sub> crystals is characterised using a microscope in dark field configuration. The samples are uniformly illuminated with unpolarised light from an incandescent light source which is tightly focused on the sample using a 100 $\times$  objective with a numerical aperture of 0.85. Scattered light is detected with a CCD camera and simultaneously collected with a multimode fibre that is attached to a

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cooled spectrometer. This fibre acts as a confocal pinhole ensuring that light is only collected from a region with a diameter of  $\sim 1 \mu\text{m}$ . The samples are precisely positioned with piezoelectric translation stages so that scattering from the edge and centre regions of the triangular MoS<sub>2</sub> crystals can be distinguished. The scattered light spectrum can be measured in a single shot, much faster than is possible with wavelength-scanned photocurrent measurements.

The scattering from MoS<sub>2</sub> crystals is compared in Figure 1 between quartz (left column) and Au (right column) substrates. Images taken with a CCD camera (Fig. 1(a)) reveal dramatically different scattering colours of the triangular MoS<sub>2</sub> crystals on the different substrates. Such triangular monolayer crystallites are typical for the hexagonal MoS<sub>2</sub> structure.<sup>2,4</sup> While scattering on quartz has a blue colour, scattering on Au is green. Strong white spots in the images correspond to areas of multiple or folded MoS<sub>2</sub> layers resulting from the growth and transfer processes.<sup>16</sup> Scattering spectra (Fig. 1(b)) are collected from the edges (red curves) or the centre (blue curves) of many different MoS<sub>2</sub> crystals. In general, the scattered light intensity for MoS<sub>2</sub> on quartz is twice as strong as for MoS<sub>2</sub> on Au. In both systems, light is scattered more strongly from the edges of the monolayer than from their centre, however the spectral shape is not altered. Scattering from different crystallites varies only minimally showing a high level of consistency across all samples. In both systems, the two *K*-excitonic states *A* and *B*, split in energy due to strong spin-orbit coupling, are clearly visible as peaks at 663 nm and 620 nm. Comparing MoS<sub>2</sub> on quartz and gold substrates, we observe blue-shifts of the *A* and *B* exciton of 2.8 meV and 12.9 meV, respectively, which are however not the main focus of this paper (see below).

The apparent colour of the material in optical images is instead dominated by strong scattering at higher energies compared to the *K*-excitonic resonances.

At these shorter wavelengths, the resonant scattering amplitude is approximately four times as strong as scattering from the *K*-excitons. For a quartz substrate, only one scattering peak is seen, centred at 439 nm for light scattered from the edge of the crystal. Light scattered from the centre is shifted to the blue by 13 nm. By contrast, for MoS<sub>2</sub> on gold, a second strong scattering peak (labelled *C'*) appears at 502 nm giving the crystals a green appearance. Recent theoretical calculations<sup>8,10</sup> suggest that these resonances arise from near the gamma point, where the electronic bands become parallel thus providing a van-Hove singularity with excitonic features. We hence refer to these resonances as *C*-excitons (although their origin remains under further discussion). The *C*-excitons have a six-fold symmetry in *K*-space close to the  $\Gamma$ -point and are strongly localised in real space with calculated radii of 30 Å<sup>8</sup> to 50 Å.<sup>10</sup> In contrast, the *A* and *B* excitons at the *K*-point extend over 65 Å<sup>8</sup> to 100 Å,<sup>10</sup> but possess internal angular-momentum with a spatially separated electron and hole.

A suggestive interpretation of these results is now presented, before comparing to existing theories as well as additional experiments. We propose that the splitting of the *C*-exciton observed on gold substrates is due to the interaction with its image exciton within the metal. The 350 meV red-shift of the *C'*-exciton can be estimated from the interaction of a dipole-emitter with its reflected field from the Au surface. The frequency shift  $\Delta\omega_s$  is estimated from the complex dielectric functions of Au ( $\epsilon_{Au}$ ) and MoS<sub>2</sub> ( $\epsilon$ ) which is the emitter's environment,<sup>43</sup>

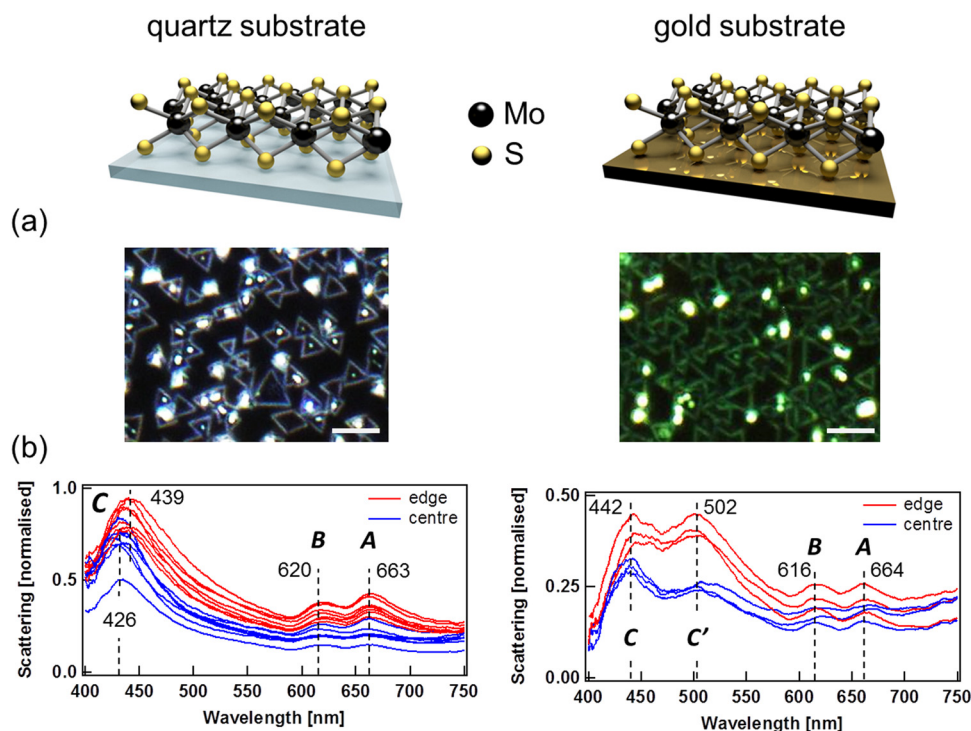


FIG. 1. Light scattering from MoS<sub>2</sub> crystals on quartz (left column) and on gold (right column) substrates. (a) Dark field images of MoS<sub>2</sub> on both substrates; Scale bars are 4  $\mu\text{m}$ . (b) Scattering spectra from the edge (red) and centre (blue) of different monolayer crystals. Data are normalised to the maximum light scattering amplitude of the quartz sample. Resonances are denoted by *A*, *B*, *C*, and *C'*.

$$\Delta\omega_s = -\frac{3}{32}q b_\infty d^{-3} \Re\left(\frac{\varepsilon_{Au} - \varepsilon}{\varepsilon_{Au} + \varepsilon}\right).$$

Here,  $q$  is the quantum yield of the emitter (around 10%),  $b_\infty$  is its decay rate (for which we use the 24 nm linewidth of the  $C$ -exciton on quartz), and  $d$  is the distance of the emitter from the surface. Estimating the complex in-plane dielectric function for MoS<sub>2</sub>,<sup>9</sup> we find frequency shifts of 350 meV for  $d = 0.6$  nm which corresponds well to the thickness of a monolayer of MoS<sub>2</sub>.<sup>41</sup> This indeed suggests that the  $C'$ -exciton arises from exciton-exciton coupling of a  $C$ -exciton in MoS<sub>2</sub> with its image in the gold surface leading to the formation of a “mirror-biexciton.”

A further comparison can be made with bandstructure calculations of bilayers, which resemble the exciton-image exciton system. Scattering spectra on quartz and gold substrates are compared with calculated absorption spectra (Fig. 2). The dashed lines are theoretical calculations including Coulomb interactions for a monolayer and bilayer of MoS<sub>2</sub> in vacuum, respectively.<sup>8</sup> Since the calculations (as typical) do not correctly predict the absolute bandgap, these spectra are rigidly shifted to lower energies by 0.3 eV to match the spectral positions of **A** and **B** excitons. While the position of the  $C$ -exciton differs in theory and experiment by 150 meV, their spectral shapes are similar. Both theory and experiment for the isolated monolayer MoS<sub>2</sub> agree that no resonance appears between the **B**- and  $C$ -excitons. In contrast, the bilayer MoS<sub>2</sub> calculation shows a second resonance around 2.5 eV ( $C'$ ) which is the result of interlayer coupling.<sup>8</sup> This suggests that indeed the mirror substrate can effectively act as an additional monolayer that couples excitons together. Alternative explanations based on doping of the MoS<sub>2</sub> sheet can be discounted since the  $K$ -excitons are not seen to be strongly affected by the substrate type, while similar results are observed if an additional insulating BN monolayer separates the Au and MoS<sub>2</sub>. We believe that  $K$ -excitons do not form mirror biexcitons because of their different angular and spin symmetry (Fig. 2(b)). The **A**-exciton possesses orbital angular momentum resulting in a repulsive interaction with the image in the surface. In contrast, the  $C$ -exciton being close to the  $\Gamma$ -point does not have angular momentum so that this configuration leads to an attractive electromagnetic force and coupling between exciton and image.

To further investigate the effect of gold in close proximity to monolayers of MoS<sub>2</sub>, the samples on both quartz and

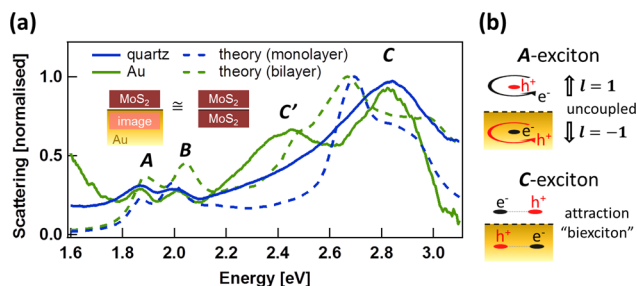


FIG. 2. (a) Comparison of experimental and theoretical scattering spectra. The blue and green curve show scattering spectra from MoS<sub>2</sub> measured on a quartz and Au substrate (insets). Theoretical calculations are shown in dashed blue for a monolayer and dashed green for a bilayer of MoS<sub>2</sub>. Resonance positions are highlighted by **A**, **B**, **C**, and  $C'$ . (b) Visualisation of **A**- and  $C$ - exciton symmetries at a mirror.

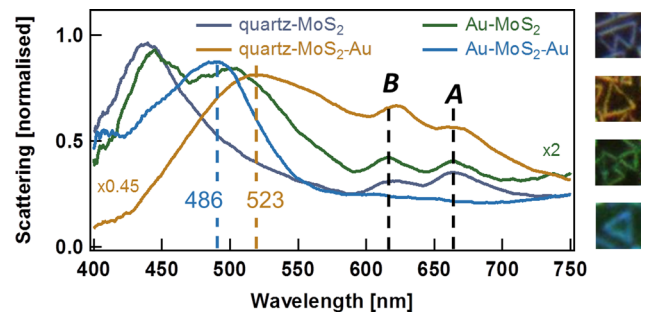


FIG. 3. Comparison of scattering spectra for: quartz-MoS<sub>2</sub>, quartz-MoS<sub>2</sub>-Au, Au-MoS<sub>2</sub>, and Au-MoS<sub>2</sub>-Au. The  $5 \times 5 \mu\text{m}$  unprocessed optical images on the right show the real colour scattered by each system.

Au substrates are coated with an additional layer of 15 nm Au. This coating is thin enough to allow for optical spectroscopy of the semiconductor through the Au and thick enough to form a smooth continuous film which is confirmed by SEM. A comparison of the scattering spectra (Fig. 3) shows the origin of the drastically different colours observed (spectra are colour coded corresponding to the raw optical images on the right). In addition to the scattering spectra of MoS<sub>2</sub> on quartz (dark-blue) and on Au (green) discussed above, the new spectra show modified features when additional Au is coated on top.

For the quartz-MoS<sub>2</sub>-Au system, both excitons **A** and **B** remain clearly visible, with resonance positions that match those in the uncoated system. In contrast, the strong scattering peak at 439 nm of the uncoated system is now shifted to the red around 523 nm. Measurements instead taken through the backside of the sample (through the quartz substrate, not shown) exhibit peaks at 435 nm and 465 nm, similar to the Au-MoS<sub>2</sub> system. On the other hand, coating the Au-MoS<sub>2</sub> with an extra layer of gold strongly diminishes both **A** and **B** exciton peaks, and the high-energy resonances shift slightly to the blue and change in intensity ratio. We speculate that the disappearance of the  $K$ -excitons occurs because the field orientation in the Au-sandwich is now perpendicular to the MoS<sub>2</sub> sheet, although screening of excitons close to the Au surface may also play a role (see supplementary material).<sup>38</sup> The overall spectral shape of the Au sandwiched MoS<sub>2</sub>, however, resembles calculated absorption spectra for bulk MoS<sub>2</sub> crystals in vacuum.<sup>38</sup> Hence, this suggests that gold layers at the top and bottom of MoS<sub>2</sub> monolayers act as mirrors that lead to multiple  $C$ -exciton-image interactions, thus mimicking bulk crystals. The proximity of free-electron metals to the tightly bound excitons in these monolayer semiconductors is thus able to create new composite particles. The resulting bound states can be useful for devices and are highly relevant for optoelectronics in these systems. Moreover, they offer the prospect that if the electron density in the substrate is gated, the exciton energies can be dynamically tuned.

In conclusion, we have shown that the optical scattering spectra of MoS<sub>2</sub> monolayers are strongly affected by gold in close proximity to the crystal. Whereas excitons at the direct band gap are little affected, the  $C$ -exciton develops a new resonance. The resonance shift is explained by coupling of the  $C$ -exciton with its image within the gold surface. Estimates of the frequency shift of an emitter close to a metallic surface agree with experimental observations. Furthermore, placing



monolayers of MoS<sub>2</sub> on a gold substrate forms the optical equivalent to MoS<sub>2</sub> bilayers due to the exciton-image coupling. Further *ab initio* calculations examining the hybridisation of the MoS<sub>2</sub> excitons with the Au will be needed to fully understand this striking effect. The work opens up the prospect for tuning optical properties of two-dimensional semiconductors with thin metallic coatings.

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