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# Design of dual-frequency electromagnetic wave absorption by interface modulation strategy

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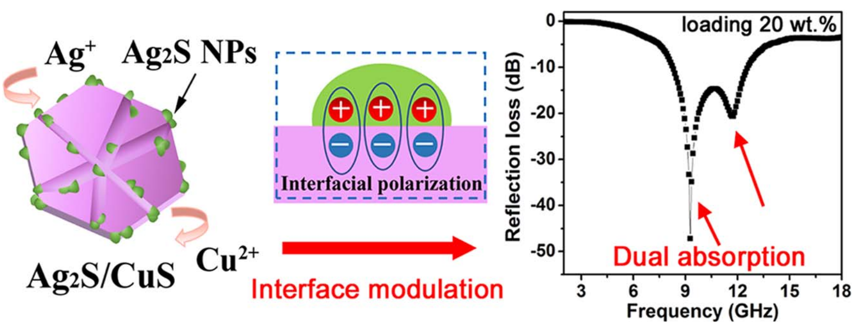
## GRAPHICAL ABSTRACT

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## ABSTRACT

Design of highly efficient electromagnetic wave (EMW) absorbing materials with continuous dual-absorption peaks is a hot topic but a challenge in anti-electromagnetic interference and pollution fields. Herein, we developed an interfacial modulation strategy to achieve this goal in dielectric nanocomposite and clarified the interface function and relevant mechanism. Experimentally, $CuS/Ag_2S$ composite with tunable heterointerface was fabricated by decorating $Ag_2S$ nanoparticles on CuS flakes via cation-exchange reaction. By changing the $Ag_2S$ loading, the heterointerface between $Ag_2S$ and CuS induced strong dielectric resonant peak and improved impedance matching, which resulted in the significant enhancement of EMW absorption and the appearance of continuous dual-absorption peaks in $CuS/Ag_2S$ composite. With a rather low filler loading of 20 wt% and a thin layer thickness of 2.89 mm, the optimized $CuS/Ag_2S$ composite exhibited continuous dual-absorption peaks with reflection losses of −47.2 and −20.6 dB, respectively. The proposed interfacial modulation strategy may potentially be used to fabricate other highly efficient dual-absorption absorbers.

## 1. Introduction

Electromagnetic wave (EMW) absorbing materials capable of converting EM energy to thermal energy have aroused considerable attention for anti-electromagnetic interference and pollution [1–3]. The rapid developments of electronic devices and wireless communication require new-generation EMW absorbing materials to possess strong absorption in a broad bandwidth with advantages of lightweight and small thickness, which are significant for practical applications [4–6]. To fulfill these requirements, intensive efforts have been devoted to the rational design and fabrication of magnetic-dielectric composite absorbers mainly through improving the impedance matching by balancing permittivity and permeability [7]. In comparison with single component absorber, the enhancement of EMW absorption has been

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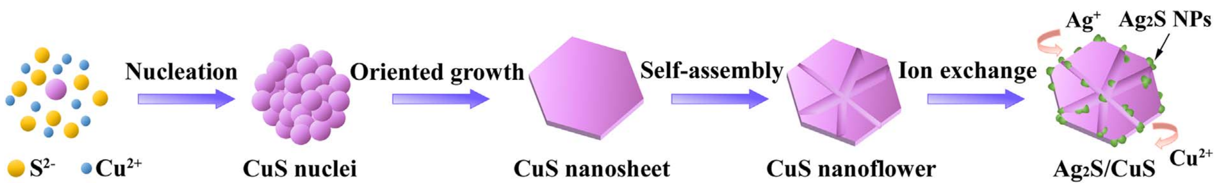


**Scheme 1.** Schematic illustration of synthesis of CuS/$Ag_2S$ composite.

achieved in various dual-loss composite absorbers such as CoNi/$TiO_2$ [8], $Fe_3O_4$/$MnO_2$ [9], Ni/$SnO_2$ [10], Fe/C [11,12], Co/CNTs [13], FeSiAl/CNTs [14], Co/C [15], and so on.

In addition to the enhanced EMW absorption, it is attractive to find that some magnetic-dielectric composite absorbers possess dual-absorption peaks which can extend the effective absorption bandwidth (reflection loss RL ≤ −10 dB). For example, Sun et al. observed separated dual EMW absorption peaks at 5.2 and 17.0 GHz in $Fe_3O_4$/graphene composite, and the effective absorption bandwidth covered 4.5 GHz [16]. Likewise, the Ni/graphene composite with separated dual-absorption peaks located at ~3.5 and ~12.5 GHz possessed stronger EMW absorption and broader effective absorption bandwidth compared with Ni nanocrystals with one absorption peak [17]. Furthermore, Cao's group recently reported a continuous dual-absorption phenomenon in Ca-substituted $BiFeO_3$ absorber that consisted of ferroelectric and ferromagnetic two phases [18]. Compared with the separated dual-absorption peaks, the continuous dual-absorption peaks are more ideal for broadband EMW absorption. They attributed the improved EMW absorbing performance to the modulation of permittivity and permeability induced by tailoring the phase boundary via hetero-atom doping [18]. Although the advantage of dual-absorption peaks has been realized, it is still challenging to develop appropriate approaches for achieving the continuous dual absorption peaks, because most EMW absorbers only possess one effective absorption peak.

In this work, we developed an interface modulation strategy to achieve enhanced EMW absorption and continuous dual-absorption peaks simultaneously. Unlike the commonly reported magnetic/dielectric hybrid EMW absorbers, we designed dielectric composite absorber to exclude the influence of magnetic-dielectric synergistic effect on EMW absorption which is helpful for clarifying the interface effect. Recently, semiconducting CuS material has been considered as a promising candidate for advanced EMW absorber due to its excellent dielectric property. Therefore, we selected representative CuS flakes to construct composite absorbers by in-situ growing $Ag_2S$ nanoparticles (NPs) on flakes through cation-exchange reaction (as illustrated in Scheme 1). The interface area between the two phases could be well adjusted by precisely controlling the $Ag_2S$ loading. It is well known that EMW absorbing performance is determined by permittivity and permeability of materials which closely associate with morphology, composition and microstructure. Due to these complicated factors, understanding the interface effect on EMW absorbing performance becomes a hot topic but a challenge. Fortunately, the design of CuS/$Ag_2S$ composite simplifies this issue because the entire dielectric compositions could exclude the influence of magnetic-dielectric synergistic effect, and meanwhile there is no morphology, geometry or defect-induced permittivity variation in this composite as the $Ag_2S$ amount changes. Our results indicated that the modulation of heterointerfaces could considerably enhance the EMW absorption of the composite, and lead to the appearance of dual-absorption peaks. The CuS/$Ag_2S$ composite with the optimum interface area exhibited continuous dual-absorption peaks with minimum RL values of −47.2 dB (at 9.28 GHz) and −20.6 dB (at 11.68 GHz) as the filler loading was only 20 wt% and layer thickness was below 3 mm. The excellent EMW absorbing performance was attributed to the significantly improved dielectric loss and impedance matching induced by interface modulation. The proposed strategy and relevant mechanism provide novel opportunities for designing highly efficient dual-absorption absorbers.

## 2. Experimental

### 2.1. Materials

Copper nitrate trihydrate ($Cu(NO_3)_2·3H_2O$, 99%), silver nitrate ($AgNO_3$, 99.8%), sulphur, cetyl trimethyl ammonium bromide (CTAB), ethylene glycol were purchased from Xilong Chemical Co., Ltd. All reagents were of analytical grade and used without further purification.

### 2.2. Preparation of flower-like CuS

The flower-like CuS was synthesized by solvothermal method. Typically, 1.8 mmol of $Cu(NO_3)_2·3H_2O$, 3.6 mmol of sulphur and 0.456 mmol of CTAB were dissolved in 200 mL of ethylene glycol through sonication. Subsequently, the solution was kept at 90 °C for 2 h under magnetic stirring to create a stable solution. The obtained solution was then transferred to Teflon-lined stainless-steel autoclaves, and maintained at 140 °C for 3 h. After cooling to room temperature, the product was collected by centrifugation, washed with water and ethanol for several times, and dried in vacuum overnight.

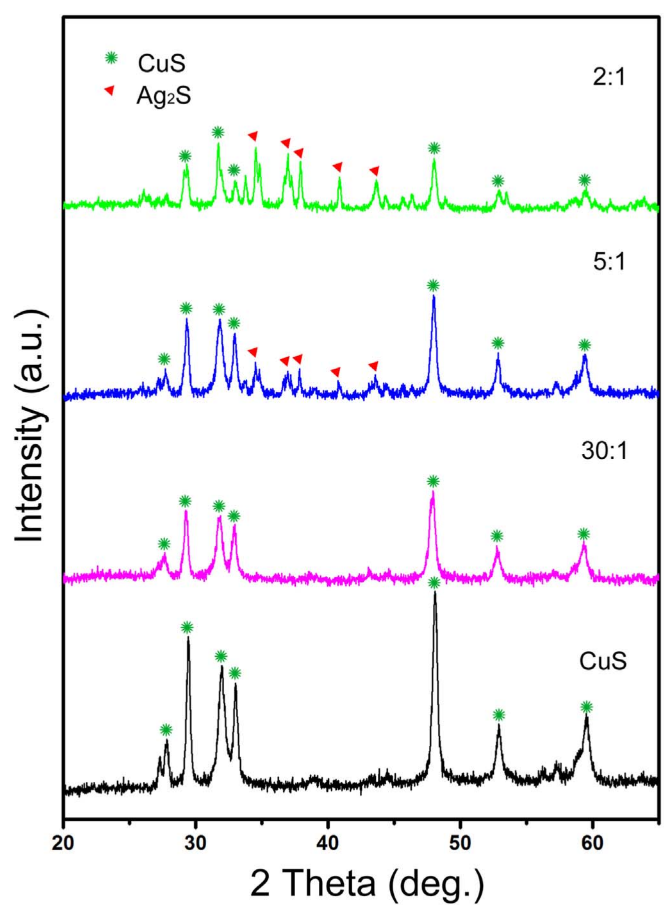


**Fig. 1.** XRD patterns of CuS and CuS/$Ag_2S$ composites with different mole ratios of Cu:Ag.