

Supplementary Materials

Tuning terahertz wave absorption in diamond/NdFeB composite films via magnetic interfacial engineering

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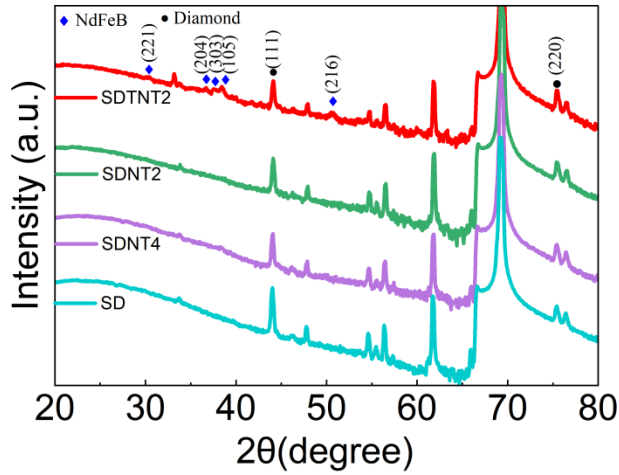
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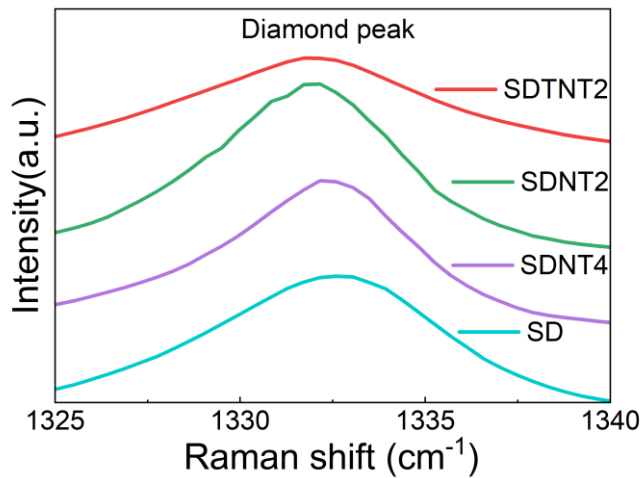
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Supplementary Figure 1. X-ray diffraction (XRD) was performed with a geometry of x-rays incident perpendicularly to the film surface, using Cu K α radiation.

In the diffraction pattern, besides the characteristic diffraction peaks of the diamond and NdFeB phases, the characteristic signal of the Si is also observed, which appears as the strongest peak at $2\theta = 69^\circ$. The Si (001) substrate was selected for diamond deposition considering epitaxial growth feasibility, process compatibility, cost, and substrate size^[1]. Single-crystal silicon offers the largest available wafer sizes at low cost, and bias-enhanced nucleation enables heteroepitaxial growth of diamond on Si (001) with high nucleation density^[2]. During initial growth, a silicon carbide buffer layer forms preferentially on the silicon surface, effectively relieving lattice mismatch and thermal stress, thus providing a compatible template for subsequent high-quality diamond growth^[3].



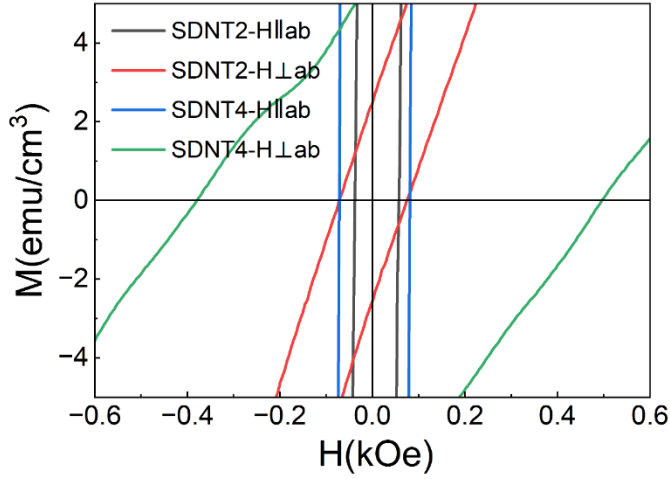
Supplementary Figure 2. Enlarged view of the Raman spectra in the 1320-1340 cm^{-1}

range, comparing the diamond peaks of different samples.

Supplementary Table 1. Raman spectral characteristic parameters of diamond/NdFeB composite thin films, including the peak position, and full width at half maximum (FWHM) of the characteristic Raman peaks.

Sample	Position (cm ⁻¹)	FWHM (cm ⁻¹)
SDTNT2	1331.95541	11.29
SDTN2	1331.67813	10.31
SDTN4	1331.94748	7.49
SD	1332.1892	8.17

Raman spectrum shows a broadened diamond peak accompanied. These observations point to the occurrence of interfacial interactions during film deposition and subsequent thermal processes^[4]. The observed amorphization and the formation of sp² carbon can be rationalized by the following thermodynamic and kinetic considerations. Carbon atoms in the diamond lattice (sp³ covalent network) and those in the metallic NdFeB environment reside in entirely different chemical states. The chemical potential of carbon in the metal is substantially lower than that in diamond, as carbon atoms can form stronger C-M (M = Fe, Nd) hybrid bonds or interstitial solid solutions with transition metals^[5]. This chemical potential gradient provides the primary driving force for carbon migration from diamond into the NdFeB layer. In the directly contacted state, the interface possesses extremely high energy due to the substantial lattice mismatch (about 146%) and the difference in bonding nature. Carbon diffusion and subsequent amorphization form a transition layer that relieves lattice strain and lowers the interfacial energy, providing an additional thermodynamic driving force^[6,7]. Furthermore, the unoccupied 3d orbitals of Fe atoms can interact with the 2p_z electrons of carbon, facilitating C-C bond cleavage and reconstruction. This catalytic effect lowers the kinetic barrier for the sp³ → sp² transformation, promoting the conversion of diamond-derived carbon into a more stable sp² configuration at the interface, and supports the proposed role of interfacial engineering in enhancing the terahertz absorption performance^[8,9].



Supplementary Figure 3. Magnified view of the central region of the M – H loops for the SDNT2 and SDNT4 samples, showing the coercivity (H_c) behavior.

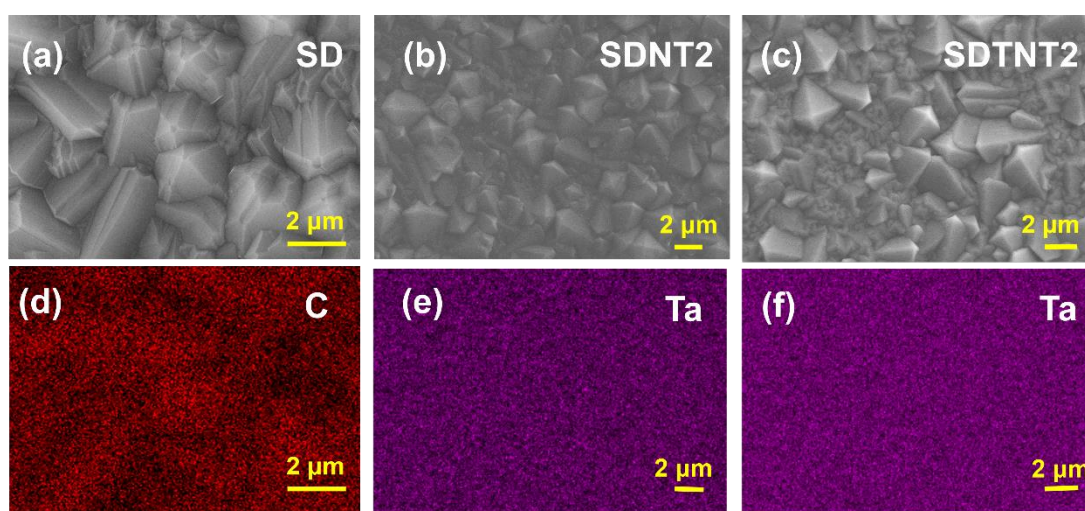
Supplementary Table 2. In-plane and out-of-plane coercivity of diamond/NdFeB composite thin films

Sample	H_c (Oe) in plane	H_c (Oe) out-of-plane
SDTNT2	14170.44	13226.28
SDTN2	57.52	74.64
SDTN4	81.04	496.27

In the SDTNT2 sample, the Ta interlayer effectively suppresses the diffusion of carbon from diamond into the NdFeB layer, so that the $\text{Nd}_2\text{Fe}_{14}\text{B}$ hard magnetic phase maintains its intact tetragonal crystal structure and high uniaxial magnetocrystalline anisotropy, exhibiting typical bulk-like hard magnetic characteristics. In contrast, the SDNT2 and SDNT4 samples lack the Ta interlayer, and carbon atoms from diamond disrupt the long-range ordered structure of $\text{Nd}_2\text{Fe}_{14}\text{B}$, leading to amorphization^[10]. The magnetic moment orientation of amorphous NdFeB is mainly governed by short-range order and shape anisotropy, showing soft magnetic properties.

For the SDNT2 and SDNT4 samples, the in-plane coercivity is smaller than the out-of-plane coercivity, indicating that the in-plane direction is the easy magnetization direction. This is because amorphization eliminates magnetocrystalline anisotropy, and shape anisotropy becomes the dominant factor determining the easy axis. The demagnetizing factor in the in-plane direction is extremely small, and the magnetic

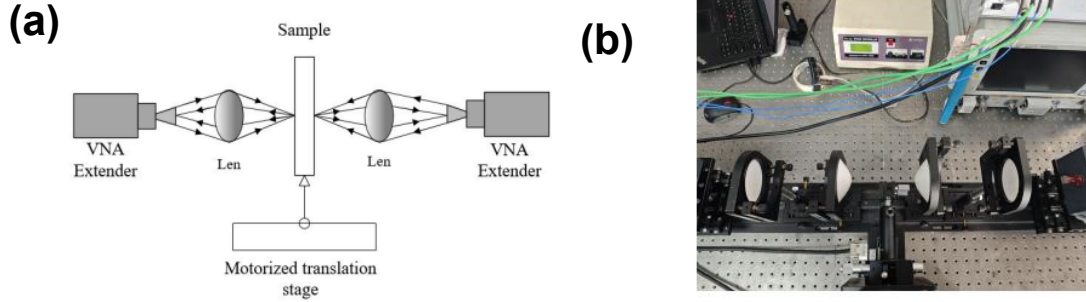
moments tend to lie within the film plane to minimize magnetostatic energy. Therefore, in-plane magnetization is easier, and the coercivity is lower. The SDTNT2 sample shows the opposite trend, indicating that the easy axis is perpendicular to the film plane. This originates from the fact that the c -axis of crystalline $\text{Nd}_2\text{Fe}_{14}\text{B}$ is the easy magnetization axis, and in this sample, the c -axis tends to grow along the film normal, forming a perpendicular magnetocrystalline anisotropy^[11]. The magnetocrystalline anisotropy energy of the crystalline sample is much larger than the shape anisotropy energy of the thin film, so the easy axis direction is dominated by magnetocrystalline anisotropy.



Supplementary Figure 4. SEM morphology and corresponding EDS elemental mapping of the samples. (a, d) SD: surface morphology and C elemental distribution. (b, e) SDNT2: surface morphology and Ta elemental distribution. (c, f) SDTNT2: surface morphology and Ta elemental distribution.

Supplementary Table 3. Surface roughness of diamond/NdFeB composite thin films

Sample	R_a (nm)	R_{max} (nm)
SDTNT2	89.4	752
SDTN2	89.7	783
SDTN4	98.1	844
SD	114	982

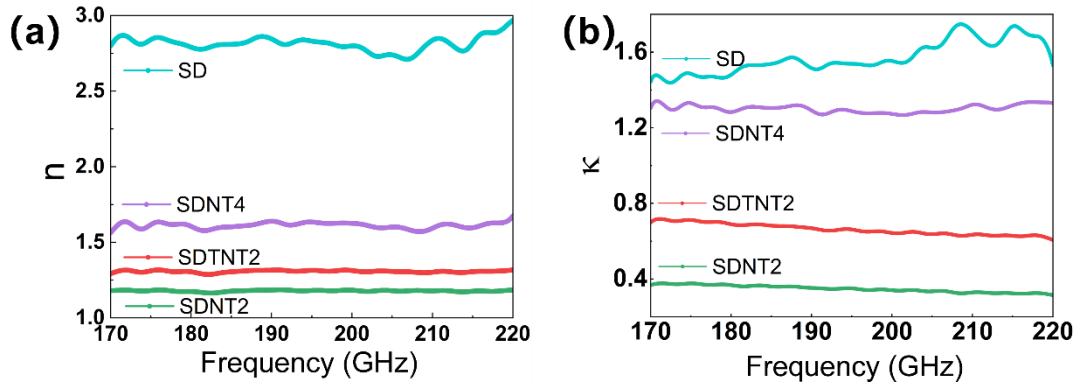


Supplementary Figure 5. Schematic and photographic images of the VNA measurement system. (a) Schematic diagram of the free-space transmission/reflection measurement setup; (b) photographic image of the actual testing configuration used for S -parameter acquisition over the 170-220 GHz frequency range.

The vector network analyzer (VNA) system directly measures S -parameters (such as S_{11} and S_{21}) of the sample. These S -parameters do not directly provide the complex permittivity or complex permeability of the material. To extract these intrinsic electromagnetic parameters, an inversion procedure based on transmission/reflection theory, such as the classical Nicolson-Ross-Weir (NRW) method, is required to retrieve them from the measured S -parameters. The measured S -parameters are first converted into the complex reflection coefficient (Γ) and complex transmission coefficient (T). Subsequently, based on the transmission/reflection theory, the complex refractive index ($N = n - i\kappa$) is extracted from Γ and T . The complex permittivity $\epsilon = \epsilon' - i\epsilon''$ and permeability $\mu = \mu' - i\mu''$ are related by $\epsilon = N^2/\mu$. For non-magnetic materials, it is a widely adopted convention in VNA analysis to assume $\mu' = 1$ and $\mu'' = 0$, thereby enabling the direct determination of the complex permittivity from the refractive index. This approximation is applicable to the vast majority of dielectric materials and composite materials in the terahertz range^[12].

In our case, the ferromagnetic resonance frequency of NdFeB-based materials falls in the GHz range (approximately 10-25 GHz), far below the 170-220 GHz band, resulting in a negligible μ'' and hence negligible resonance-type magnetic loss^[13]. This assumption is further supported by the amorphized state of the NdFeB layer in the directly contacted sample (SDNT2), which exhibits negligible magnetocrystalline anisotropy and therefore an even weaker high-frequency magnetic response. Hysteresis loss is also minimal due to the extremely low coercivity of the amorphized

NdFeB layer and is ineffective at THz frequencies. Eddy current loss is effectively suppressed by the thin-film geometry, as the film thickness is far smaller than both the skin depth and the wavelength scale at 170-220 GHz^[14]. In addition to conventional magnetic loss, the NdFeB layer may offer an ultrafast spin-related dissipation channel via spin-orbit coupling (e.g., spin pumping or ultrafast demagnetization)^[15,16]. However, diamond lacks strong spin-orbit coupling for efficient spin-to-charge conversion, while amorphized NdFeB introduces abundant spin-scattering centers that rapidly dissipate spin currents. Therefore, the contribution of such spin-related mechanisms is substantially smaller than that of dielectric loss in this work. Consequently, the observed absorption is predominantly governed by electric-field-driven dielectric loss mechanisms. Accordingly, following the standard protocol for such materials in THz-TDS studies, we set $\mu' = 1$ and $\mu'' = 0$ throughout the permittivity extraction procedure.



Supplementary Figure 6. Refractive properties of diamond/NdFeB composite thin films in the frequency range of 170-220 GHz. (a) real part of refractive constant, (b) imaginary part of refractive constant.

Supplementary Table 4. Comparison of the diamond/NdFeB composite film with representative THz absorbers reported in the literature

Material system	Frequency	Absorptivity	Thickness	Ref.
Graphene	THz	intrinsically low		[17]
Graphene/PI/Au	0.3-1.3 THz	60%-99%	165 μm	[17]
Ti ₃ C ₂ T _x MXene	0.5-4.5 THz	< 50%	10.2 nm	[18]
MXene/FeNi composite	THz band	75%	0.6 mm	[19]

Metallic metamaterial absorber	1-3 THz	>90% (<5% bandwidth)		[20]
Diamond/NdFeB (SDNT2)	170-220 GHz	47%-70%	0.5 mm	This work

The central contribution of this work is not to compete with the highest-performance terahertz absorbers reported to date, but rather to demonstrate a conceptual shift: transforming diamond into an effective absorber through interfacial engineering with NdFeB, while simultaneously improving impedance matching via magnetic modulation. In other words, this study focuses on endowing diamond with a new functionality rather than pursuing peak absorptivity for its own sake.

To provide a clearer context for this approach, a comparative analysis with representative terahertz absorbers in the literature is presented. As summarized in Supplementary Table 4, pristine graphene and ultrathin MXene films exhibit intrinsically low absorption (< 50% in most cases), whereas graphene/PI/Au multilayers and MXene/FeNi composites can achieve higher absorptivity (up to 75-99%) at the cost of complex multilayer architectures, additional magnetic decoration, and increased thickness. Metallic metamaterial absorbers, though capable of high peak absorption, are inherently narrowband (< 5% bandwidth) and rely on precision lithography. By contrast, the SDNT2 sample developed in this work achieves broadband absorption of 47-70% in the 170-220 GHz band through a simple CVD and magnetron sputtering process without photolithography. While its absorptivity may not be the highest among all reported THz absorbers, the key novelty lies in offering a new material paradigm that combines simplicity, scalability, and reasonable performance, transforming diamond into an active absorbing component.

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