Electronic Supplementary Material (ESI) for Materials Horizons.

Metal Dimer Nanojunctions Composites with Magnetic Materials for Magnetic Field Sensing

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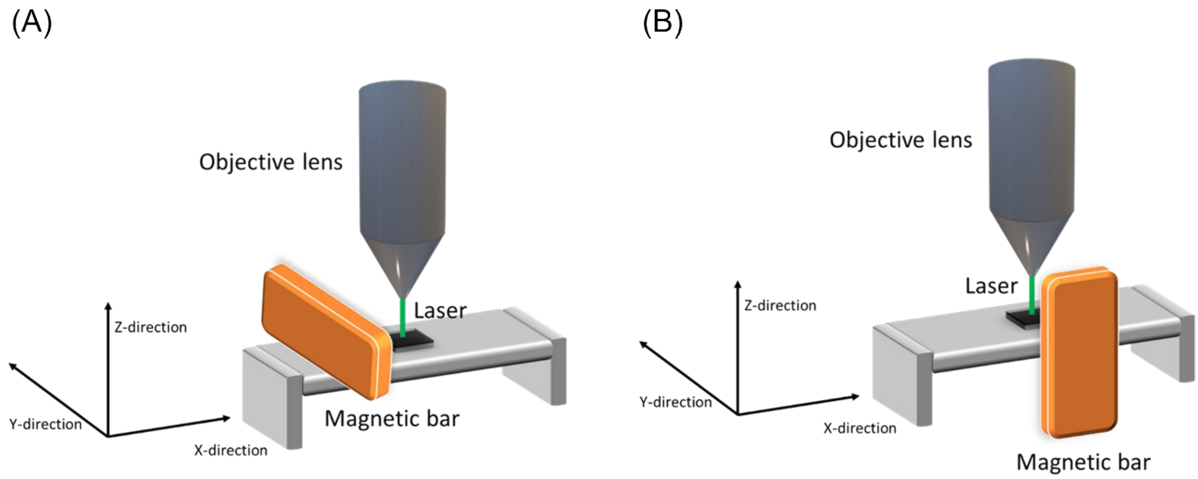


Figure S1 Experimental setup for measurement the Raman spectra of dimer nanojunctions composite under different directions of the external magnetic force. (A) y-axis magnetic field: magnetic field parallel to the substrate; (B) z-axis magnetic field: magnetic field perpendicular to the substrate.

Table S1 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with naphthalenediamine under external magnetic field (parallel or perpendicular to the substrate).

	Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I' - I_0)/I_0$ (%)			
		y-axis		z-axis	
Naphthalenediamine (NDA)	1358 C-N stretching	485.4	507.4 ± 31.1	128.5	138.3 ± 13.7
	1594 N-H deformation vibration	529.4		148.0	
PS	1450 CH_2 scissoring	528.8	581.3 ± 74.2	226.3	205.8 ± 29.1
	1615 Ring-skeletal stretch	633.7		185.2	
PVP	1327 C-N stretching / CH_2 wagging	505.6	534.9 ± 33.8	204.9	270.6 ± 113.7
	1501 C-N stretching	527.1		204.9	
	1760 C=O stretching	571.8		401.9	

Table S2 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with naphthalenediamine under external magnetic field (parallel or perpendicular to the substrate).

	Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I' - I_0)/I_0$ (%)			
		y-axis		z-axis	
Naphthalenediamine (NDA)	1358 C-N stretching	723.4	688.2 ± 52.6	168.1	166.7 ± 2.09
	1594 N-H deformation vibration	649.0		165.2	
PS	1450 CH_2 scissoring	654.8	744.7 ± 127.2	202.8	182.2 ± 29.1
	1615 Ring-skeletal stretch	834.7		161.7	
PVP	1327 C-N stretching / CH_2 wagging	723.4	771.6 ± 133.2	202.5	205.2 ± 15.7
	1501 C-N stretching	669.2		222.1	
	1760 C=O stretching	922.1		191.0	

Table S3 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with benzimidazole under external magnetic field (parallel or perpendicular to the substrate).

	Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	204.6	224.5 ± 27.6	566.1	596.5 ± 43.1
	1595 N-H deformation vibration	244.0		626.7	
PS	1450 CH_2 scissoring	187.3	179.0 ±11.3	543.6	598.5 ± 7.1
	1615 Ring-skeletal stretch	171.8		652.8	
PVP	1327 C-N stretching / CH_2 wagging	231.3	160.7±88.6	452.8	546.0 ± 186.7
	1501 C-N stretching	166.1		760.8	
	1760 C=O stretching	097.4		424.4	

Table S4 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with benzimidazole under external magnetic field (parallel or perpendicular to the substrate).

	Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	747.1	684.0 ± 89.2	881.4	702.9 ± 252.4
	1595 N-H deformation vibration	621.0		524.4	
PS	1450 CH_2 scissoring	638.9	714.9 ± 107.6	861.7	759.8 ± 144.1
	1615 Ring-skeletal stretch	791.0		657.9	
PVP	1327 C-N stretching / CH_2 wagging	577.0	558.7 ± 293.7	758.5	722.4 ± 430.7
	1501 C-N stretching	842.8		1133.9	
	1760 C=O stretching	256.3		274.7	

Table S5 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with different naphthalenediamine concentrations under external magnetic field (parallel or perpendicular to the substrate).

		Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
			y-axis		z-axis	
Naphthalene diamine (NDA)	High conc.	1358 C-N stretching	485.4	507.4 ± 31.1	128.5	138.3 ± 13.7
		1594 N-H deformation vibration	529.4		148.0	
	low conc.	1358 C-N stretching	289.6	269.1± 28.9	138.4	119.8 ± 26.3
		1594 N-H deformation vibration	248.7		101.3	
PS	High conc.	1450 CH_2 scissoring	528.8	581.3 ± 74.2	226.3	205.8 ± 29.1
		1615 Ring-skeletal stretch	633.7		185.2	
	low conc.	1450 CH_2 scissoring	267.3	266. 9 ± 0.64	122.0	114.2 ± 10.9
		1615 Ring-skeletal stretch	266.4		106.5	
PVP	High conc.	1327 C-N stretching / CH_2 wagging	505.6	534.9 ± 33.8	204.9	270.6 ± 113.7
		1501 C-N stretching	527.1		204.9	
		1760 C=O stretching	571.8		401.9	
	low conc.	1327 C-N stretching / CH_2 wagging	268.0	252.5± 22.7	120.9	99.6±18.9
		1501 C-N stretching	226.5		84.7	
		1760 C=O stretching	262.9		931.4	

Table S6 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with different naphthalenediamine concentrations under external magnetic field (parallel or perpendicular to the substrate).

		Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
			y-axis		z-axis	
Naphthalene diamine (NDA)	High conc.	1358 C-N stretching	723.4	688.2 ± 52.6	168.1	166.7 ± 2.09
		1594 N-H deformation vibration	649.0		165.2	
	low conc.	1358 C-N stretching	244.2	292.0 ± 67.6	93.2	112.0± 26.6
		1594 N-H deformation vibration	339.8		130.8	
PS	High conc.	1450 CH_2 scissoring	528.8	581.3 ± 74.2	226.3	205.8 ± 29.1
		1615 Ring-skeletal stretch	633.7		185.2	
	low conc.	1450 CH_2 scissoring	245.6	253.7± 11.4	108.9	121.8± 18.2
		1615 Ring-skeletal stretch	261.7		134.7	
PVP	High conc.	1327 C-N stretching / CH_2 wagging	505.6	534.9 ± 33.8	204.9	270.6 ± 113.7
		1501 C-N stretching	527.1		204.9	
		1760 C=O stretching	571.8		401.9	
	low conc.	1327 C-N stretching / CH_2 wagging	251.1	272.6 ± 19.2	111.0	107.6 ±12.2
		1501 C-N stretching	278.4		117.8	
		1760 C=O stretching	288.2		94.1	

Table S7 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with different benzimidazole concentrations under external magnetic field (parallel or perpendicular to the substrate).

		Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
			y-axis		z-axis	
Benzimidazole (BIM)	High conc.	1265 (C-H) in-plane bending	204.6	224.5 ± 27.6	566.1	596.5 ± 43.1
		1595 N-H deformation vibration	244.0		626.7	
	low conc.	1265 (C-H) in-plane bending	184.9	164.5 ± 27.6	229.5	256.7± 38.5
		1595 N-H deformation vibration	145.9		284.0	
PS	High conc.	1450 CH_2 scissoring	187.3	179.0 ±11.3	543.6	598.5 ± 7.1
		1615 Ring-skeletal stretch	171.8		652.8	
	low conc.	1450 CH_2 scissoring	180.9	147.0 ± 46.7	454.2	422.8± 44.4
		1615 Ring-skeletal stretch	114.1		391.5	
PVP	High conc.	1327 C-N stretching / CH_2 wagging	231.3	160.7± 88.6	452.8	546.0 ± 186.7
		1501 C-N stretching	166.1		760.8	
		1760 C=O stretching	097.4		424.4	
	low conc.	1327 C-N stretching / CH_2 wagging	118.9	129.3 ± 31.6	568.0	414.6 ± 198.7
		1501 C-N stretching	165.9		485.7	
		1760 C=O stretching	105.9		190.2	

Table S8 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with different benzimidazole concentrations under external magnetic field (parallel or perpendicular to the substrate).

		Chemical shift (cm^{-1})/vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)			
			y-axis		z-axis	
Benzimidazole (BIM)	High conc.	1265 (C-H) in-plane bending	747.1	684.0 ± 89.2	881.4	702.9 ± 252.4
		1595 N-H deformation vibration	621.0		524.4	
	low conc.	1265 (C-H) in-plane bending	157.0	226.0 ± 97.6	430.0	481.8 ± 73.3
		1595 N-H deformation vibration	295.1		533.7	
PS	High conc.	1450 CH_2 scissoring	638.9	714.9 ± 107.6	861.7	759.8 ± 144.1
		1615 Ring-skeletal stretch	791.0		657.9	
	low conc.	1450 CH_2 scissoring	199.6	233.2 ± 47.5	403.4	513.3 ± 155.4
		1615 Ring-skeletal stretch	266.8		623.2	
PVP	High conc.	1327 C-N stretching / CH_2 wagging	577.0	558.7 ± 293.7	758.5	722.4 ± 430.7
		1501 C-N stretching	842.8		1133.9	
		1760 C=O stretching	256.3		274.7	
	low conc.	1327 C-N stretching / CH_2 wagging	224.5	293.7 ± 63.8	462.9	450.4 ± 85.1
		1501 C-N stretching	306.5		528.6	
		1760 C=O stretching	350.3		359.8	

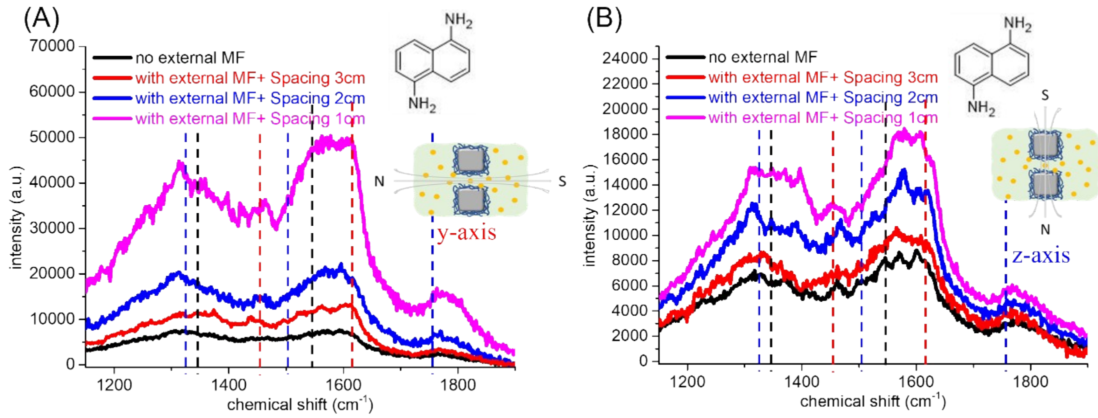


Figure S2 Raman spectra of vertical dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S9 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	485.4	507.4 ± 31.1	128.5	138.3 ± 13.7	155.7	172.4 ± 23.5	61.0	53.5 ±10.7	63.7	68.6 ± 6.9	28.1	33.7 ± 7.9
	1594 N-H deformation vibration	529.4		148.0		189.0		45.9		73.5		39.3	
PS	1450 CH_2 scissoring	528.8	581.3 ± 74.2	226.3	205.8 ± 29.1	148.1	169.4 ± 30.1	106.6	77.5 ± 41.1	65.4	79.7 ± 20.2	56.5	49.3 ± 10.2
	1615 Ring-skeletal stretch	633.7		185.2		190.7		48.4		94.1		42.1	
PVP	1327 C-N stretching / CH_2 wagging	505.6	534.9 ± 33.8	204.9	270.6 ± 113.7	176.8	188.2 ± 23.2	81.2	125.5 ± 72.0	47.3	52.0 ± 14.5	55.3	57.9 ± 13.2
	1501 C-N stretching	527.1		204.9		173.0		86.7		68.3		46.1	
	1760 C=O stretching	571.8		401.9		214.9		208.6		40.6		72.2	

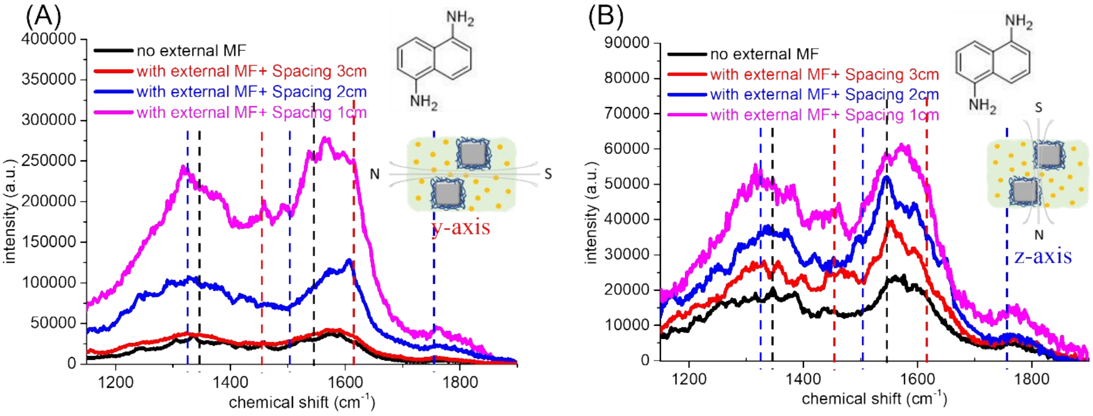


Figure S3 Raman spectra of horizontal dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S10 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Naphthalenediamine (NDA)	1358 C-N stretching	723.4	688.2 ± 52.6	168.1	166.7 ± 2.09	263.6	151.7 ± 10.0	101.3	101.5 ± 0.3	88.6	89.4 ± 1.1	51.0	49.9 ± 1.5
	1594 N-H deformation vibration	649.0		165.2		227.3		101.7		90.2		48.8	
PS	1450 CH_2 scissoring	654.8	744.7 ± 127.2	202.8	182.2 ± 29.1	227.4	103.4± 77.5	98.8	94.3 ± 6.3	54.3	94.4 ± 56.8	74.3	53.3 ± 29.6
	1615 Ring-skeletal stretch	834.7		161.7		343.1		89.8		134.6		32.4	
PVP	1327 C-N stretching / CH_2 wagging	723.4	771.6 ± 133.2	202.5	205.2 ± 15.7	259.6	128.8 ± 30.6	115.3	125.3 ± 34.0	34.6	73.7 ± 68.2	60.3	48.9 ± 29.5
	1501 C-N stretching	669.2		222.1		204.6		163.2		34.2		71.0	
	1760 C=O stretching	922.1		191.0		430.4		97.4		152.5		15.4	

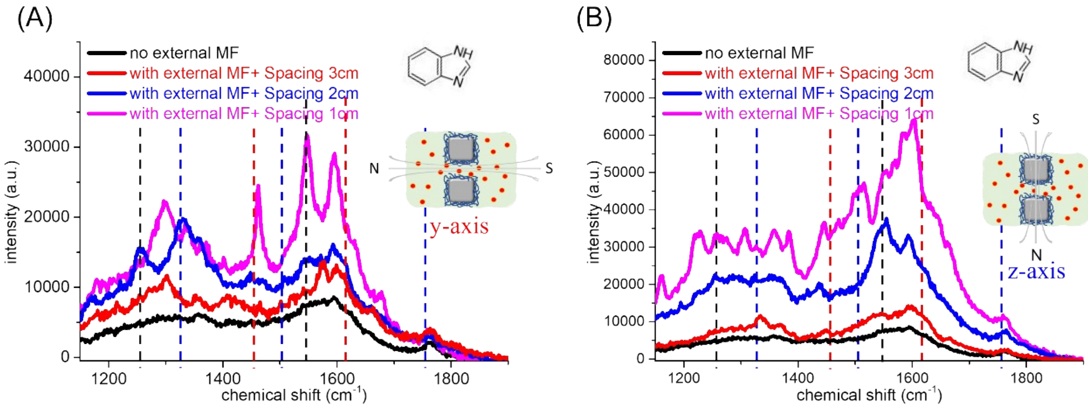


Figure S4 Raman spectra of vertical dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S11 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	204.6	224.5 ± 27.6	566.1	596.5 ± 43.1	197.7	138.5 ± 82.7	34.9	312.0 ± 39.6	85.6	64.0 ± 29.7	69.7	64.5± 6.4
	1595 N-H deformation vibration	244.0		626.7		80.5		284.1		43.2		60.2	
PS	1450 CH_2 scissoring	187.3	179.0 ±11.3	543.6	598.5 ± 7.1	117.0	108.5 ± 12.0	242.9	264.0 ± 31.1	28.2	43.5 ± 21.9	42.1	61.5 ± 27.6
	1615 Ring-skeletal stretch	171.8		652.8		100.2		286.4		59.1		81.1	
PVP	1327 C-N stretching / CH_2 wagging	231.3	160.7±88.6	452.8	546.0 ± 186.7	209.8	122.3 ± 81.2	282.0	265.3 ± 24.7	67.8	58.0 ± 18.2	90.3	59.3 ± 39.3
	1501 C-N stretching	166.1		760.8		110.5		277.2		37.7		73.6	
	1760 C=O stretching	097.4		424.4		048.3		237.9		70.2		15.4	

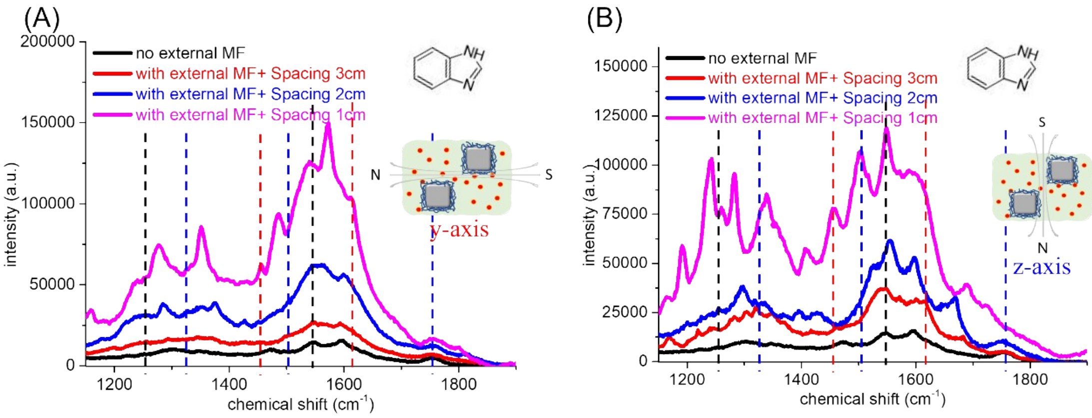


Figure S5 Raman spectra of horizontal dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S12 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	747.1	684.0 ± 89.2	881.4	702.9 ± 252.4	298.7	278.5 ± 28.6	93.3	241.0 ± 4.2	238.5	79.4 ± 19.7	127.2	112.7 ± 20.5
	1595 N-H deformation vibration	621.0		524.4		258.3		65.5		244.2		98.1	
PS	1450 CH_2 scissoring	638.9	714.9 ± 107.6	861.7	759.8 ± 144.1	258.4	294.1 ± 50.4	87.5	190.5 ± 23.3	174.0	88.2± 1.1	98.7	132.2 ± 47.5
	1615 Ring-skeletal stretch	791.0		657.9		329.7		89.0		207.4		165.8	
PVP	1327 C-N stretching / CH_2 wagging	577.0	558.7 ± 293.7	758.5	722.4 ± 430.7	284.2	268.6 ± 94.3	88.0	199.7 ± 91.2	207.9	81.5 ± 39.0	182.8	137.8 ± 43.3
	1501 C-N stretching	842.8		1133.9		354.2		116.8		287.2		134.2	
	1760 C=O stretching	256.3		274.7		167.5		39.6		105.7		9.6	

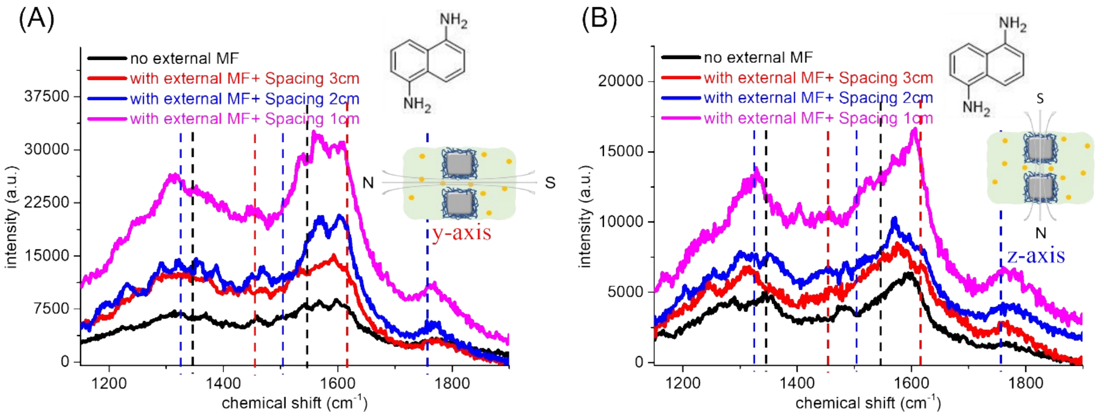


Figure S6 Raman spectra of vertical dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S13 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	289.6	269.1± 28.9	138.4	119.8 ± 26.3	118.7	118.6 ± 0.2	66.5	57.8 ± 12.3	90.0	77.1 ± 18.3	26.1	20.3 ± 8.1
	1594 N-H deformation vibration	248.7		101.3		118.5		49.1		64.1		14.6	
PS	1450 CH_2 scissoring	267.3	266. 9 ± 0.6	122.0	114.2 ± 10.9	106.5	129.0 ± 31.8	71.5	71.8 ± 0.30	70.4	68.8 ± 2.2	16.2	17.6 ± 2.0
	1615 Ring-skeletal stretch	266.4		106.5		151.5		72.0		67.2		19.0	
PVP	1327 C-N stretching / CH_2 wagging	268.0	252.5± 22.7	120.9	99.6±18.9	103.1	88.5 ± 14.2	61.5	59.3 ± 2.0	74.2	56.0 ± 19.1	21.9	20.5 ± 1.6
	1501 C-N stretching	226.5		84.7		87.9		58.7		57.6		18.8	
	1760 C=O stretching	262.9		931.4		74.7		57.6		0.00		21.0	

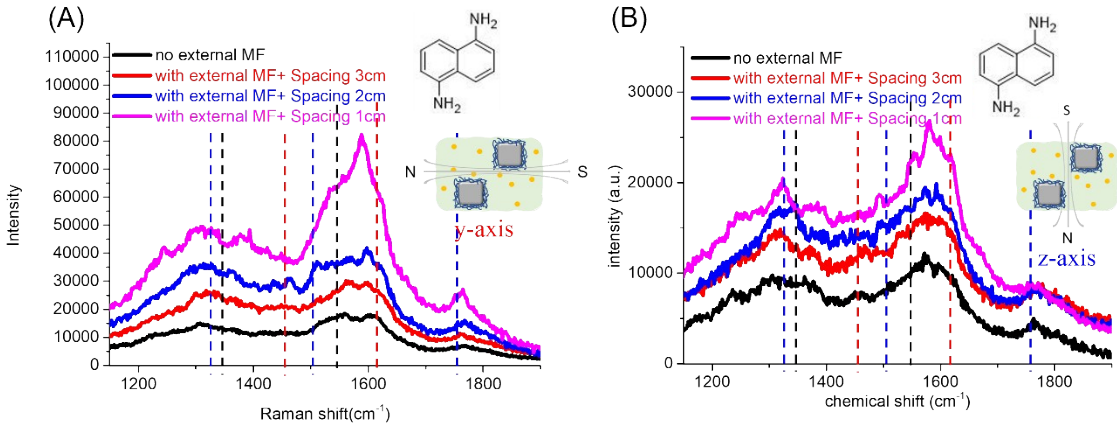


Figure S7 Raman spectra of horizontal dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S14 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	244.2	292.0 ± 67.6	93.2	112.0 ± 26.6	164.1	149.2 ± 21.1	72.8	74.0 ± 1.7	75.2	80.9 ± 8.1	42.0	44.9 ± 4.2
	1594 N-H deformation vibration	339.8		130.8		134.3		75.2		86.7		47.9	
PS	1450 CH_2 scissoring	245.6	253.7± 11.4	108.9	121.8± 18.2	144.4	127.6 ± 23.8	71.0	73.4 ± 3.4	69.0	72.6± 5.1	46.4	50.2± 5.3
	1615 Ring-skeletal stretch	261.7		134.7		110.8		75.7		76.2		54.0	
PVP	1327 C-N stretching / CH_2 wagging	251.1		111.0		163.1		82.9		60.7		51.2	
	1501 C-N stretching	278.4	272.6 ± 19.2	117.8	107.6 ±12.2	166.2	147.8± 29.3	91.4	82.3± 9.5	65.8	62.0± 3.3	53.4	58.4 ± 10.6
	1760 C=O stretching	288.2		94.1		114.0		72.5		59.6		70.6	

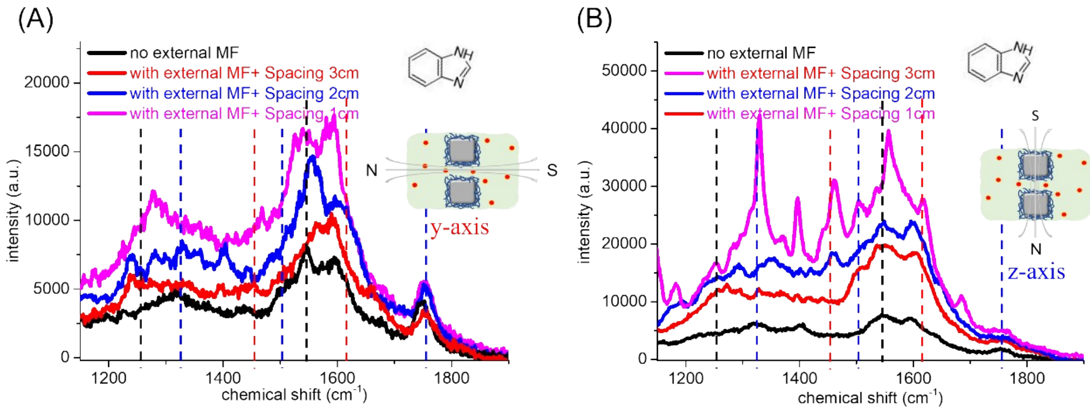


Figure S8 Raman spectra of vertical dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S15 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	184.9	164.5 ± 27.6	229.5	256.7± 38.5	64.2	68.4 ± 5.7	199.6	217.9 ± 25.8	17.2	19.5 ± 3.5	158.0	154.9 ± 4.3
	1595 N-H deformation vibration	145.9		284.0		72.6		236.1		22.7		151.9	
PS	1450 CH_2 scissoring	180.9	147.0 ± 46.7	454.2	422.8± 44.4	76.4	90.3 ± 19.8	286.8	282.2 ± 6.5	46.4	37.5 ± 12.0	133.6	170.5 ± 52.1
	1615 Ring-skeletal stretch	114.1		391.5		104.3		277.6		29.0		207.4	
PVP	1327 C-N stretching / CH_2 wagging	118.9	129.3 ± 31.6	568.0	414.6 ± 198.7	84.9	91.7 ± 7.5	175.4	198.7 ± 102.8	16.6	23.7 ± 8.0	236.3	144.4 ±79.8
	1501 C-N stretching	165.9		485.7		90.6		311.2		32.9		236.3	
	1760 C=O stretching	105.9		190.2		99.5		109.6		23.2		92.2	

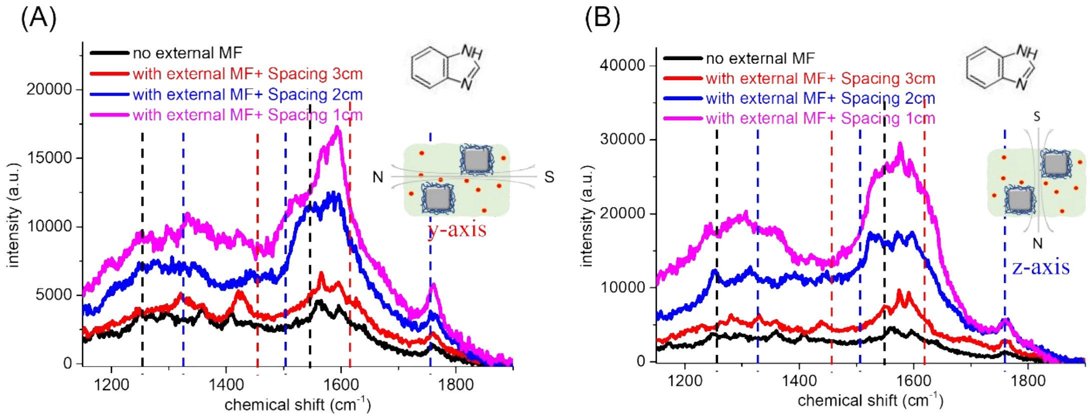


Figure S9 Raman spectra of horizontal dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by changing the spacing between the composite and the magnetic source (magnetic bar). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively.

Table S16 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force.

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
		Spacing =1 cm				Spacing =2 cm				Spacing =3 cm			
		y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	157.0	226.0 ± 97.6	430.0	481.8 ± 73.3	97.3	144.2 ± 66.3	260.1	240.0 ± 28.3	9.9	24.9 ± 21.2	53.1	75.2 ± 31.4
	1595 N-H deformation vibration	295.1		533.7		191.1		220.6		39.9		97.4	
PS	1450 CH_2 scissoring	199.6	233.2 ± 47.5	403.4	513.3 ± 155.4	138.1	154.0 ± 22.5	160.7	195.0 ± 49.5	32.6	39.9 ± 10.3	74.1	81.0± 9.7
	1615 Ring-skeletal stretch	266.8		623.2		170.0		230.5		47.2		87.9	
PVP	1327 C-N stretching / CH_2 wagging	224.5	293.7 ± 63.8	462.9	450.4 ± 85.1	131.0	153.7 ± 20.8	1730.2	200.0 ± 23.8	47.1	53.1 ± 25.5	84.1	89.4 ± 17.0
	1501 C-N stretching	306.5		528.6		171.8		218.0		31.1		75.6	
	1760 C=O stretching	350.3		359.8		158.3		209.0		81.0		108.4	

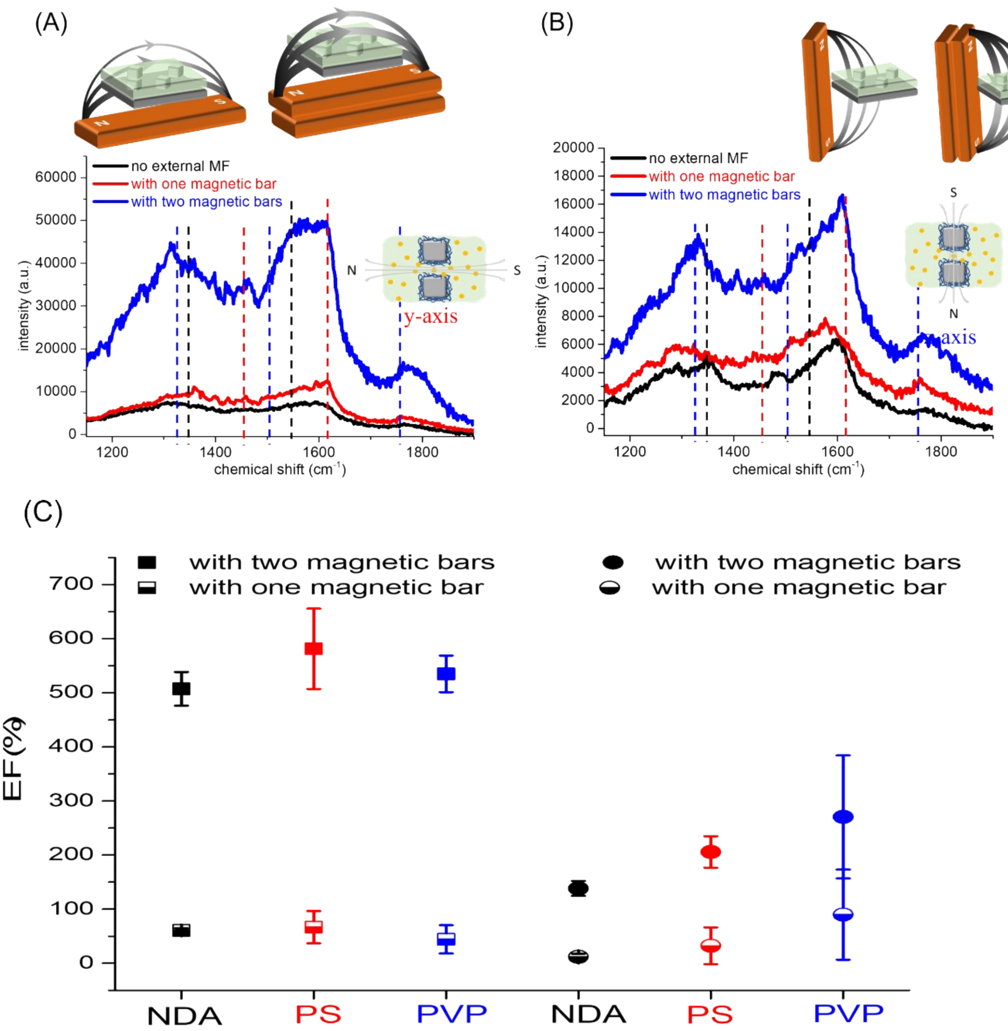
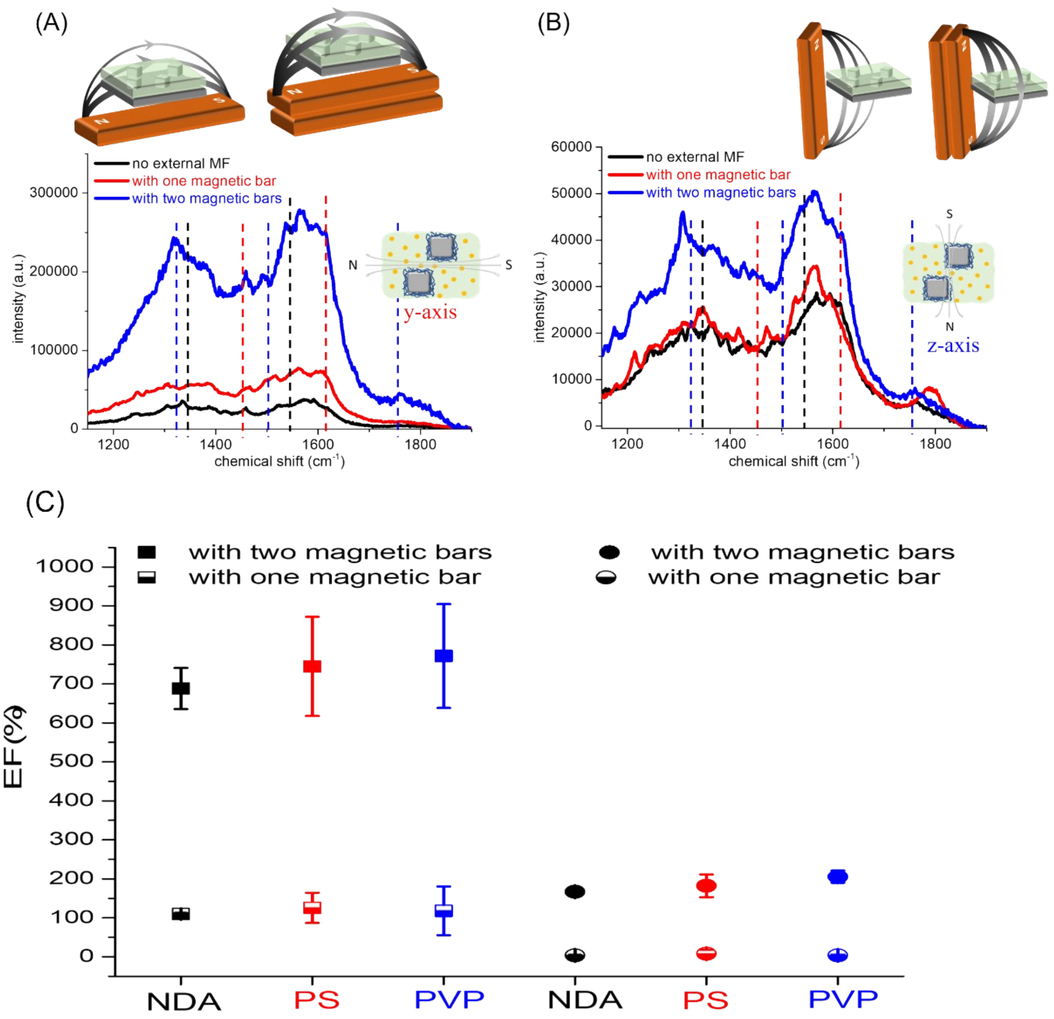


Figure S10 Raman spectra of vertical dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S17 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force (the number of magnetic bars).

<table>
<tr><th rowspan="3"></th><th rowspan="3">Chemical shift (cm^{-1})
/vibration mode</th><th colspan="8">Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)</th></tr>
<tr><th colspan="4">Two magnetic bars</th><th colspan="4">One magnetic bar</th></tr>
<tr><th colspan="2">y-axis</th><th colspan="2">z-axis</th><th colspan="2">y-axis</th><th colspan="2">z-axis</th></tr>
<tr><td rowspan="2">Naphthalene diamine (NDA)</td><td>1358
C-N stretching</td><td>485.4</td><td rowspan="2">507.4 ± 31.1</td><td>128.5</td><td rowspan="2">138.3 ± 13.7</td><td>63.3</td><td rowspan="2">60.5 ± 4.0</td><td>7.7</td><td rowspan="2">11.8 ± 5.8</td></tr>
<tr><td>1594
N-H deformation vibration</td><td>529.4</td><td>148.0</td><td>57.7</td><td>15.9</td></tr>
<tr><td rowspan="2">PS</td><td>1450
CH_2 scissoring</td><td>528.8</td><td rowspan="2">581.3 ± 74.2</td><td>226.3</td><td rowspan="2">205.8 ± 29.1</td><td>45.7</td><td rowspan="2">66.8 ± 29.8</td><td>56.3</td><td rowspan="2">32.3 ± 34.0</td></tr>
<tr><td>1615
Ring-skeletal stretch</td><td>633.7</td><td>185.2</td><td>87.8</td><td>8.3</td></tr>
<tr><td rowspan="3">PVP</td><td>1327
C-N stretching / CH_2 wagging</td><td>505.6</td><td rowspan="3">534.9 ± 33.8</td><td>204.9</td><td rowspan="3">270.6 ± 113.7</td><td>24.2</td><td rowspan="3">44.2 ± 26.1</td><td>23.3</td><td rowspan="3">89.8 ± 83.3</td></tr>
<tr><td>1501
C-N stretching</td><td>527.1</td><td>204.9</td><td>34.7</td><td>63.0</td></tr>
<tr><td>1760
C=O stretching</td><td>571.8</td><td>401.9</td><td>73.7</td><td>183.3</td></tr>
</table>



Fig

ure S11 Raman spectra of horizontal dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S18 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with naphthalenediamine under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	723.4	688.2 ± 52.6	168.1	166.7 ± 2.09	107.6	109.5 ± 2.7	4.2183	3.4± 1.1
	1594 N-H deformation vibration	649.0		165.2		111.4		2.621	
PS	1450 CH_2 scissoring	654.8	744.7 ± 127.2	202.8	182.2 ± 29.1	98.0	125.5 ± 38.8	0.8518	8.0 ± 10.1
	1615 Ring-skeletal stretch	834.7		161.7		152.9		15.17	
PVP	1327 C-N stretching / CH_2 wagging	723.4	771.6 ± 133.2	202.5	205.2 ± 15.7	53.4	117.6 ± 62.5	1.3195	3.4± 1.8
	1501 C-N stretching	669.2		222.1		178.3		4.4529	
	1760 C=O stretching	922.1		191.0		121.2		4.54	

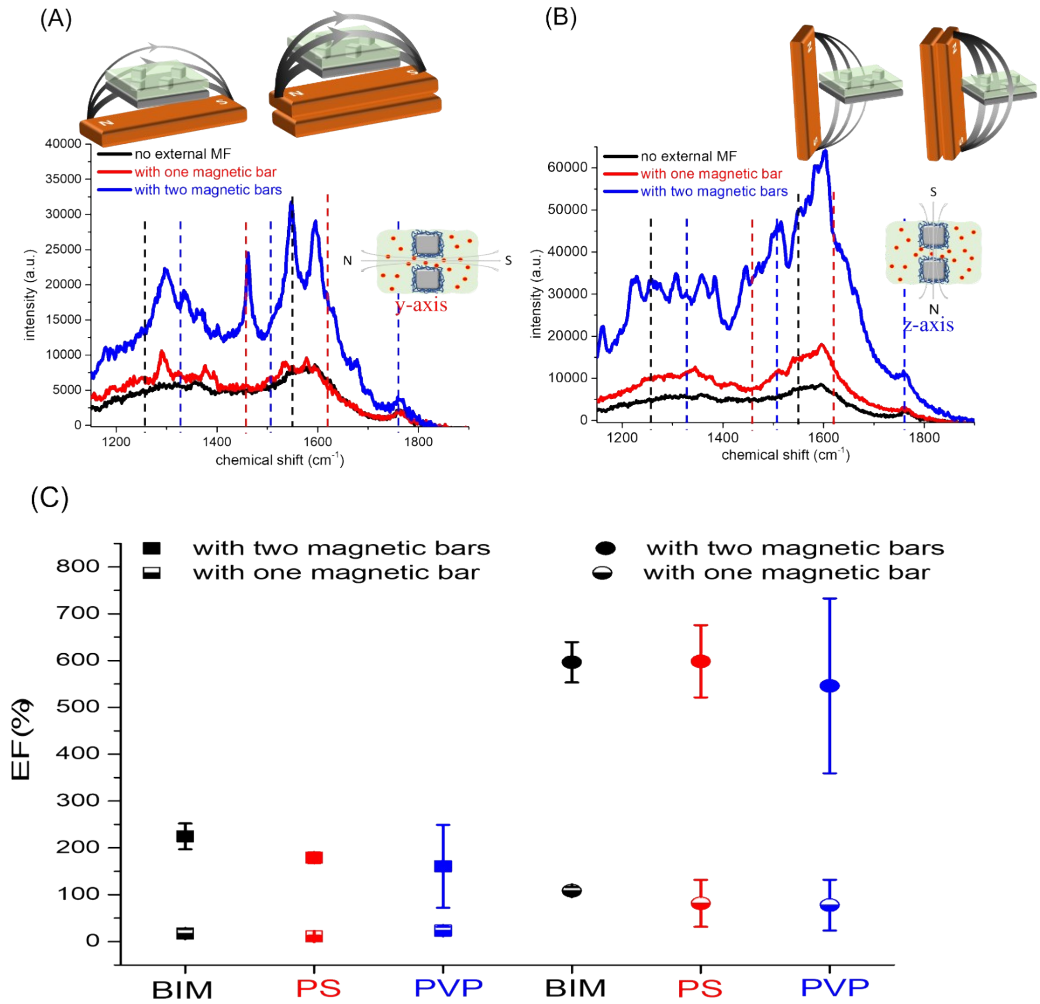


Figure S12 Raman spectra of vertical dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S19 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	204.6	224.5 ± 27.6	566.1	596.5 ± 43.1	24.1	17.3 ± 9.9	103	108.5 ± 7.8
	1595 N-H deformation vibration	244.0		626.7		10.6		114	
PS	1450 CH_2 scissoring	187.3	179.0 ±11.3	543.6	598.5 ± 7.1	12.2	11.3 ± 1.4	46	81.5 ± 50.2
	1615 Ring-skeletal stretch	171.8		652.8		10.4		117	
PVP	1327 C-N stretching / CH_2 wagging	231.3	160.7±88.6	452.8	546.0 ± 186.7	31.8	23.3 ± 8.0	109	77.7 ± 54.3
	1501 C-N stretching	166.1		760.8		24.2		109	
	1760 C=O stretching	97.4		424.4		15.8		15	

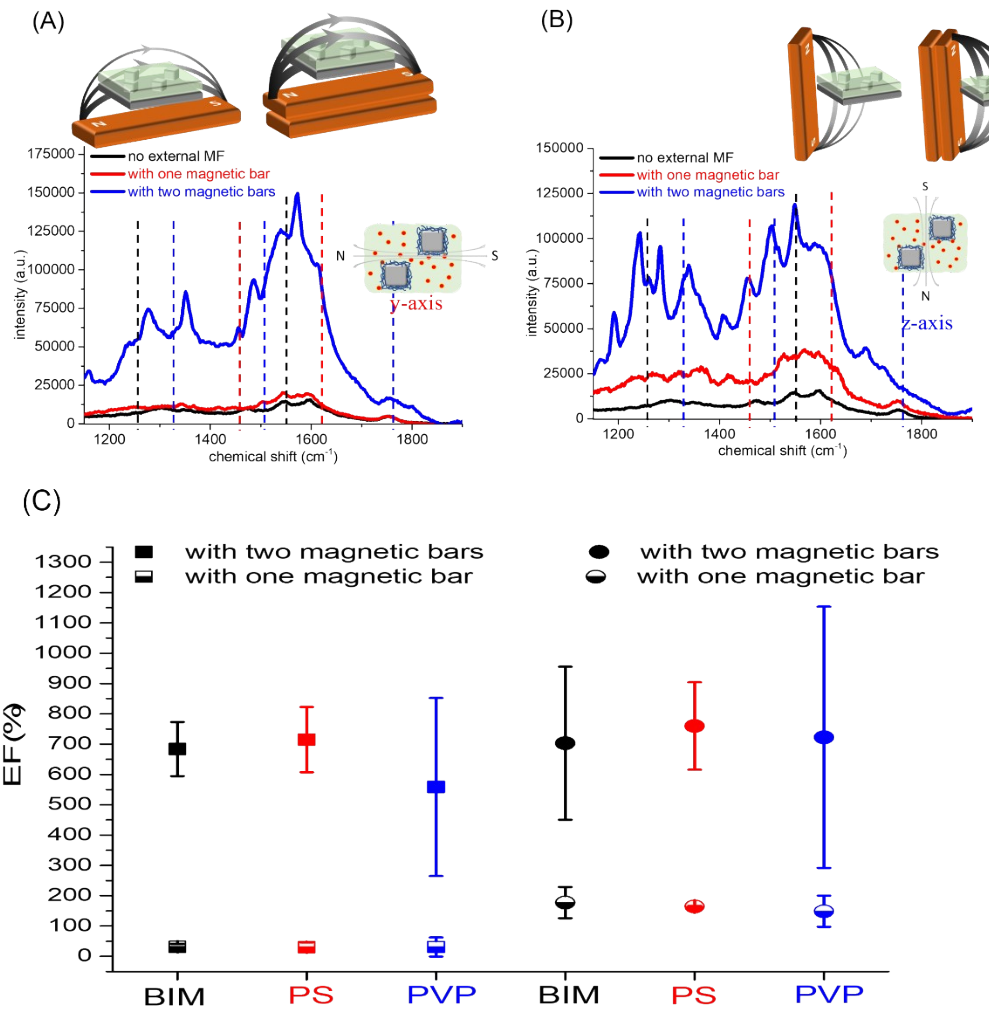


Figure S13 Raman spectra of horizontal dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S20 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with benzimidazole under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	747.1	684.0 ± 89.2	881.4	702.9 ± 252.4	38.3	31.4 ± 9.6	214	177.5 ± 51.6
	1595 N-H deformation vibration	621.0		524.4		24.6		141	
PS	1450 CH_2 scissoring	638.9	714.9 ± 107.6	861.7	759.8 ± 144.1	41.2	30.0 ± 15.8	163	164.5 ± 2.1
	1615 Ring-skeletal stretch	791.0		657.9		18.8		166	
PVP	1327 C-N stretching / CH_2 wagging	577.0	558.7 ± 293.7	758.5	722.4 ± 430.7	29.7	30.8± 31.4	156	148.7 ± 51.4
	1501 C-N stretching	842.8		1133.9		62.8		196	
	1760 C=O stretching	256.3		274.7		0.00		94	

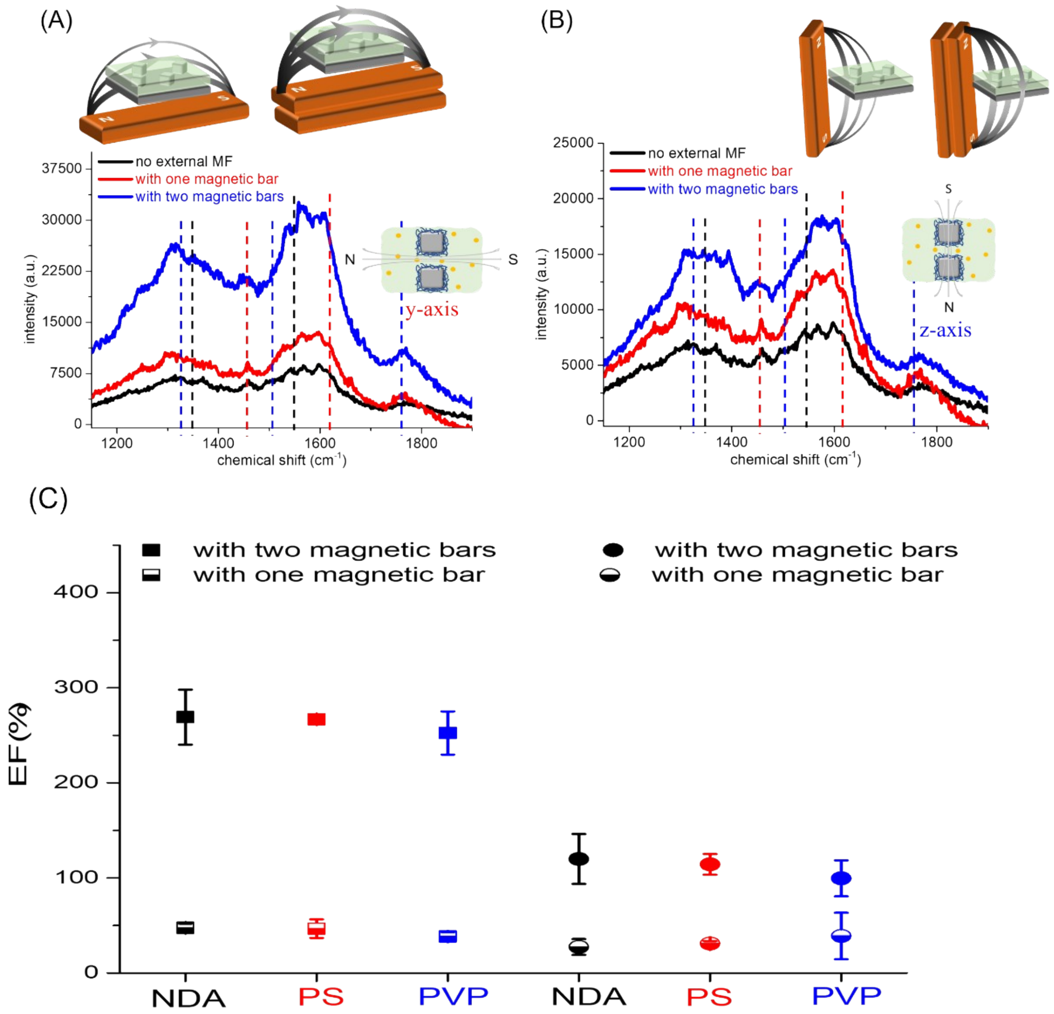


Figure S14 Raman spectra of vertical dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S21 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with half concentration of naphthalenediamine under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	289.6	269.1± 28.9	138.4	119.8 ± 26.3	44.4	47.5 ± 4.4	33.3	27.3 ± 8.4
	1594 N-H deformation vibration	248.7		101.3		50.7		21.4	
PS	1450 CH_2 scissoring	267.3	266. 9 ± 0.64	122.0	114.2 ± 10.9	39.7	46.6 ± 9.8	32.8	31.0 ± 2.5
	1615 Ring-skeletal stretch	266.4		106.5		53.6		29.2	
PVP	1327 C-N stretching / CH_2 wagging	268.0	252.5± 22.7	120.9	99.6±18.9	42.4	38.4± 4.4	23.5	38.9 ± 24.5
	1501 C-N stretching	226.5		84.7		33.6		26.0	
	1760 C=O stretching	262.9		931.4		39.2		67.1	

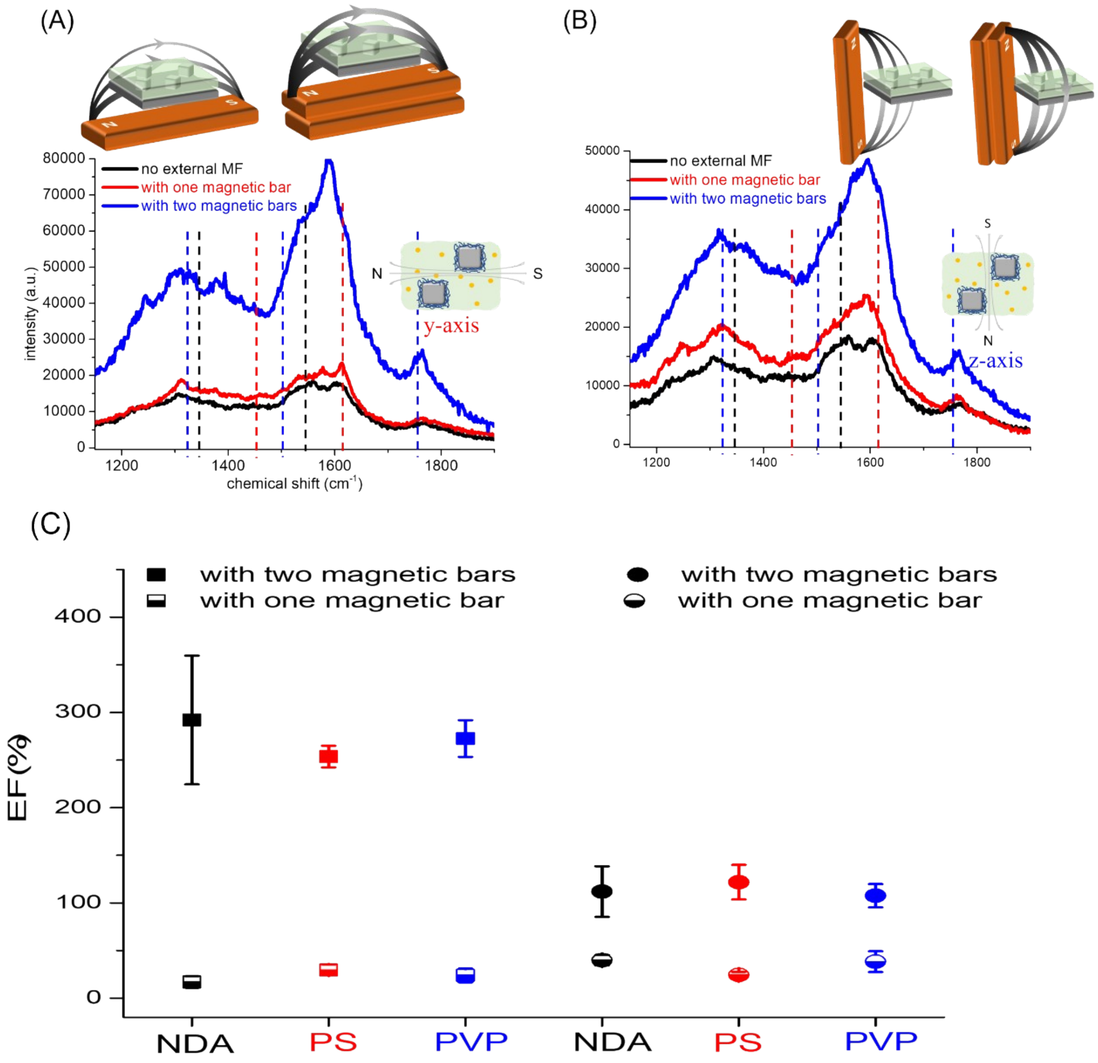


Figure S15 Raman spectra of horizontal dimer nanojunctions composite with half the concentration of naphthalenediamine under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S22 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with half concentration of naphthalenediamine under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Naphthalene diamine (NDA)	1358 C-N stretching	244.2	292.0 ± 67.6	93.2	112.0 ± 26.6	21.2	17.0 ± 6.0	36.0	39.9 ± 5.5
	1594 N-H deformation vibration	339.8		130.8		12.7		43.8	
PS	1450 CH_2 scissoring	245.6	253.7± 11.4	108.9	121.8± 18.2	25.6	29.7 ± 5.7	26.7045	24.5 ± 3.1
	1615 Ring-skeletal stretch	261.7		134.7		33.6		22.267	
PVP	1327 C-N stretching / CH_2 wagging	251.1	272.6 ± 19.2	111.0	107.6 ±12.2	18.7	23.9 ± 7.2	46.5513	38.4 ± 10.9
	1501 C-N stretching	278.4		117.8		32.1		42.6155	
	1760 C=O stretching	288.2		94.1		20.9		26.0061	

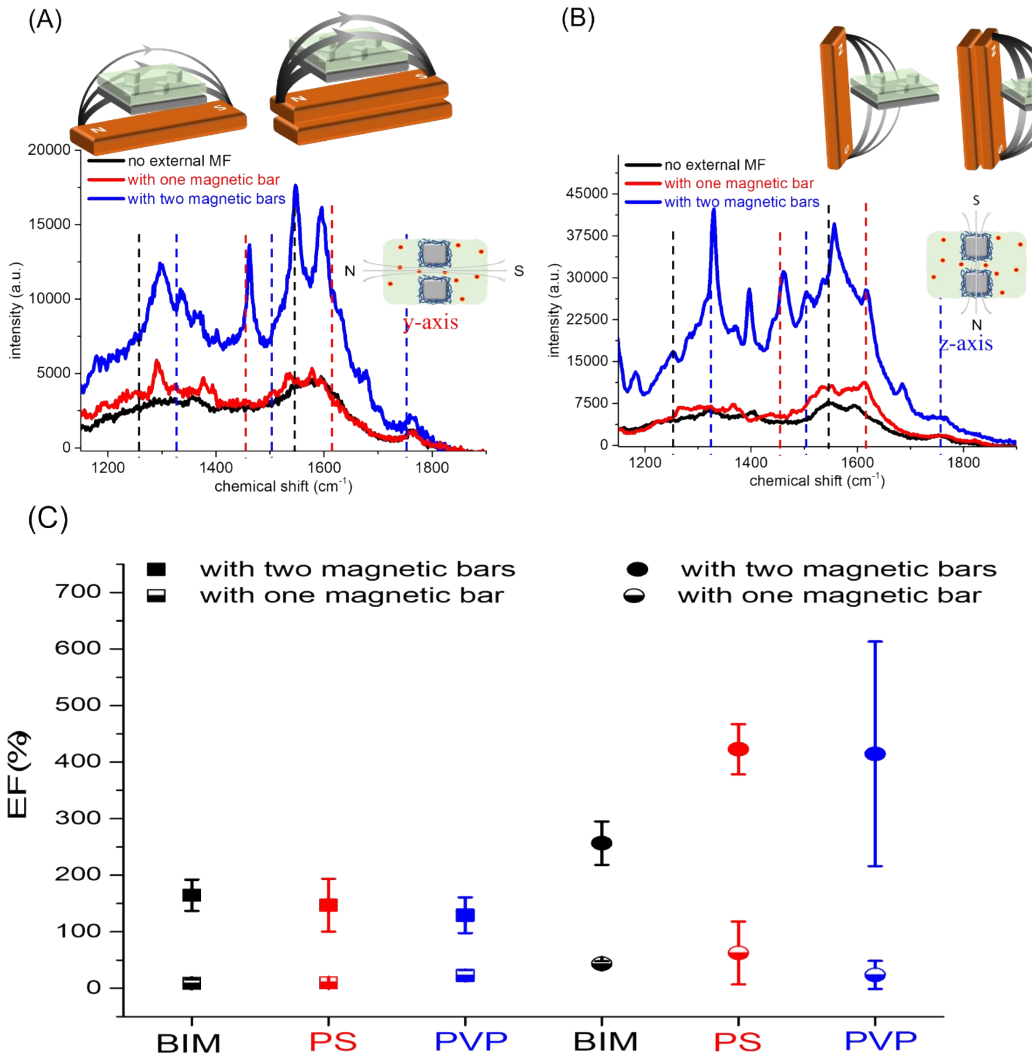


Figure S16 Raman spectra of vertical dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S23 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with half concentration of benzimidazole under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	184.9	164.5 ± 27.6	229.5	256.7± 38.5	4.3	9.1 ± 7.1	46.5	43.5 ± 4.4
	1595 N-H deformation vibration	145.9		284.0		14.0		40.4	
PS	1450 CH_2 scissoring	180.9	147.0 ± 46.7	454.2	422.8± 44.4	9.6	9.5 ± 0.7	23.2	62.6 ± 55.6
	1615 Ring-skeletal stretch	114.1		391.5		10.1		101.9	
PVP	1327 C-N stretching / CH_2 wagging	118.9	129.3 ± 31.6	568.0	414.6 ± 198.7	18.4	22.7 ± 9.9	15.5	23.7± 24.9
	1501 C-N stretching	165.9		485.7		16.0		51.6	
	1760 C=O stretching	105.9		190.2		34.7		4.0	

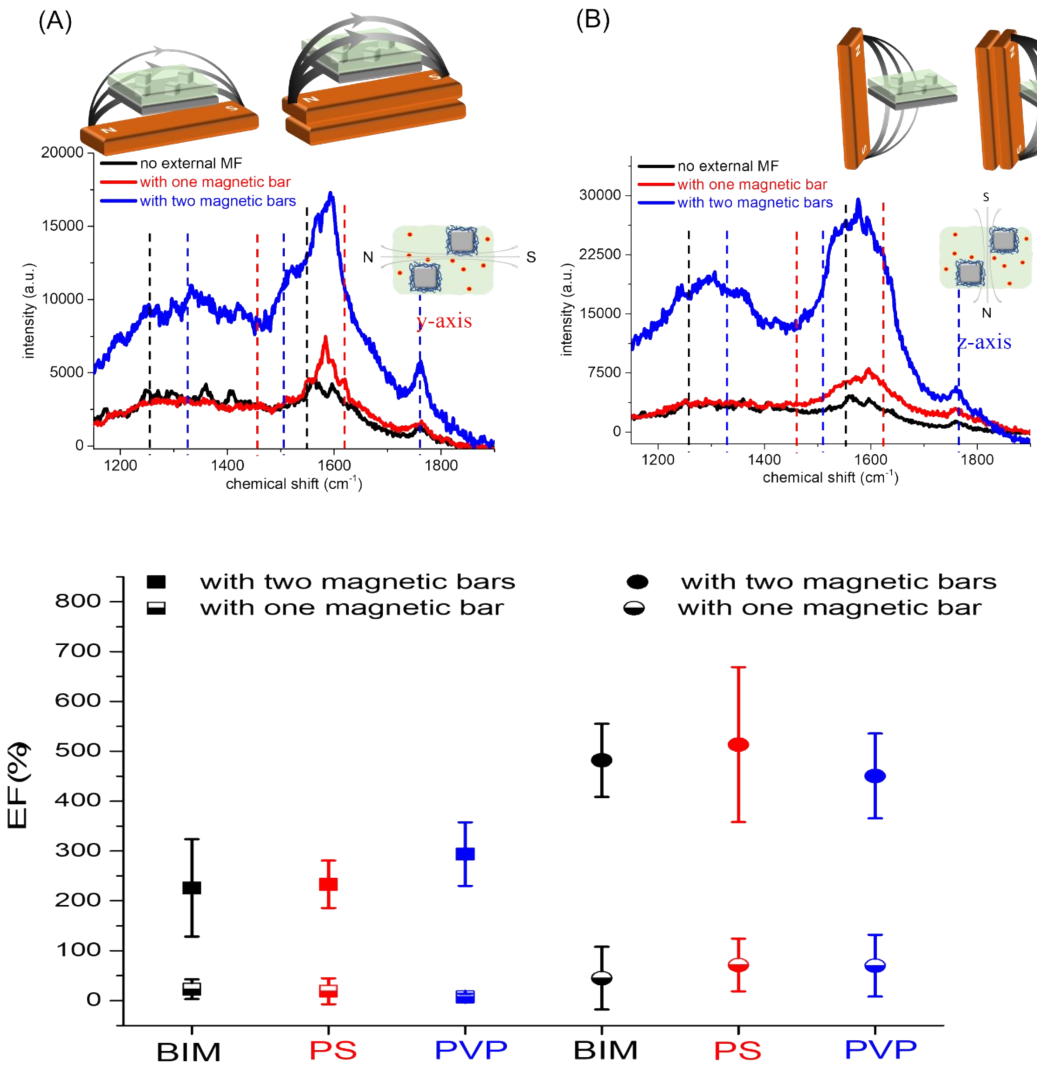
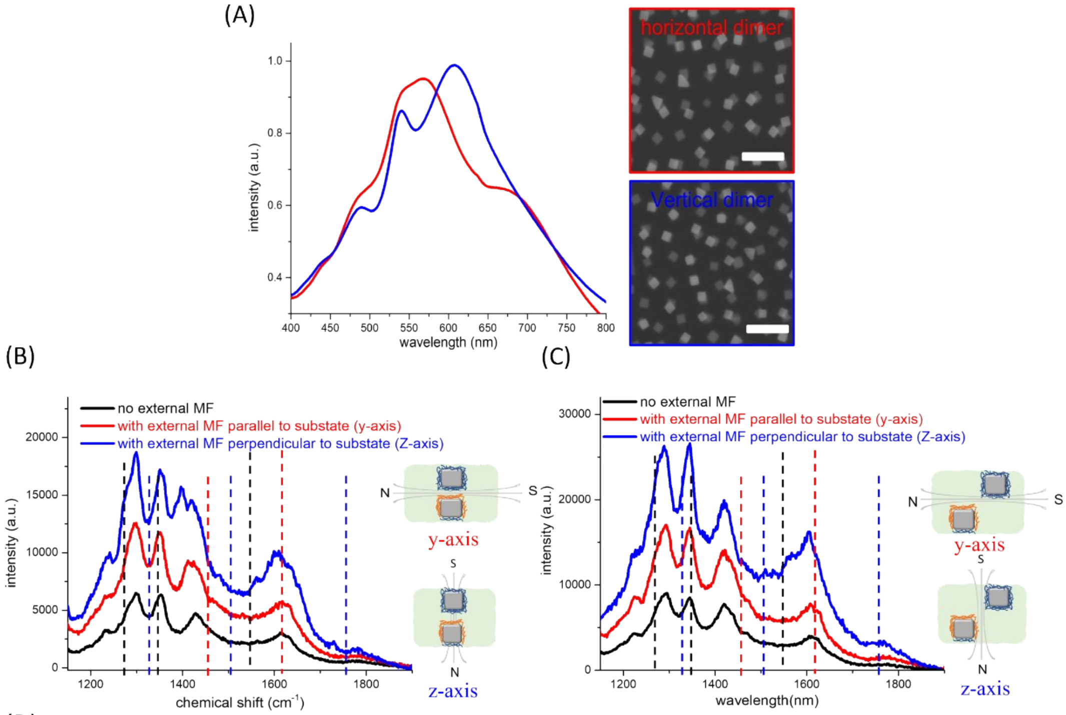


Figure S17 Raman spectra of horizontal dimer nanojunctions composite with half the concentration of benzimidazole under different strengths of the external magnetic force. The strength of the magnetic force applied on the composite can be tuned by an external magnetic source (the number of magnetic bars). Raman spectra of nanocomposites with two different morphologies of dimer nanojunctions, vertical and horizontal dimer nanojunctions, are shown in (A) and (B), respectively. The corresponding Raman signal enhancement factors of different vibration modes are shown in (C).

Table S24 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with half concentration of benzimidazole under different strengths of the external magnetic force (the number of magnetic bars).

	Chemical shift (cm^{-1}) /vibration mode	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)							
		Two magnetic bars				One magnetic bar			
		y-axis		z-axis		y-axis		z-axis	
Benzimidazole (BIM)	1265 (C-H) in-plane bending	157.0	226.0 ± 97.6	430.0	481.8 ± 73.3	8.9	22.8 ± 19.7	0.6	45.0 ± 62.8
	1595 N-H deformation vibration	295.1		533.7		36.8		89.4	
PS	1450 CH_2 scissoring	199.6	233.2 ± 47.5	403.4	513.3 ± 155.4	0.3	18.7± 26.0	34.0	71.5 ± 53.0
	1615 Ring-skeletal stretch	266.8		623.2		37.2		108.9	
PVP	1327 C-N stretching / CH_2 wagging	224.5	293.7 ± 63.8	462.9	450.4 ± 85.1	4.5	7.3 ± 6.4	19.0	70.1 ± 62.0
	1501 C-N stretching	306.5		528.6		2.7		52.2	
	1760 C=O stretching	350.3		359.8		14.5		139.1	



(D)

Chemical shift (cm^{-1})	1278	1335	1601	1450	1615	1327	1501	1760
Vibration mode	C-N-H in-plane bending	N-C-H in-plane bending	C-N stretching C=C stretching	PS CH_2 scissoring	PS Ring-skeletal stretch	PVP C-N stretching CH_2 wagging	PVP C-N stretching	PVP C=O stretch

Figure S18 Raman response of dimer-nanojunction composites with 2-Mercapto-5-nitrobenzimidazole (MNB) under external magnetic field (MF). (A) Extinction spectra of two dimer-nanojunction composites: vertical dimer (blue curve) and horizontal (red curve). The corresponding SEM images are in the right panel. (B) and (C) show the Raman spectra of vertical and horizontal dimer-nanojunction composites under three different conditions, respectively. the three different conditions are no external MF (black curve), under external MF parallel to the substrate (y-axis red curve), and under external MF perpendicular to the substrate (z-axis red curve). (D) The corresponding chemical vibration modes of MNB, PVP (ligand for AgNC), and PS (matrix) were listed.

Table S25 Enhancement factors of Raman signals (different vibration modes) of vertical dimer nanojunctions composite with different magnetic materials.

	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
	BIM (van der Waals)				NDA (hydrogen bonding)				MNB (covalent bonding)			
	y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Magnetic material	204.6	224.5 $\pm$ 27.6	566.1	596.5 $\pm$ 43.1	485.4	507.4 $\pm$ 31.1	128.5	138.3 $\pm$ 13.7	95.9	88.9 $\pm$ 7.4	192.1	189.0 $\pm$ 22.1
	244.0		626.7		529.4		148.0		81.2		165.9	
									89.6		209.9	
PS	187.3	179.0 $\pm$11.3	543.6	598.5 $\pm$ 7.1	528.8	581.3 $\pm$ 74.2	226.3	205.8 $\pm$ 29.1	88.7	97.6$\pm$ 12.6	176.0	207.3 $\pm$ 44.3
	171.8		652.8		633.7		185.2		106.5		238.6	
PVP	231.3	160.7$\pm$88.6	452.8	546.0 $\pm$ 186.7	505.6	534.9 $\pm$ 33.8	204.9	270.6 $\pm$ 113.7	96.3	87.0 $\pm$ 26.6	174.8	175.2 $\pm$ 30.6
	166.1		760.8		527.1		204.9		107.6		206.0	
	97.4		424.4		571.8		401.9		57.0		144.8	

Table S26 Enhancement factors of Raman signals (different vibration modes) of horizontal dimer nanojunctions composite with different magnetic materials.

	Enhancement factor (EF) = $(I'-I_0)/I_0$ (%)											
	BIM (van der Waals)				NDA (hydrogen bonding)				MNB (covalent bonding)			
	y-axis		z-axis		y-axis		z-axis		y-axis		z-axis	
Magnetic material	747.1	684.0 $\pm$ 89.2	881.4	702.9 $\pm$ 252.4	723.4	688.2 $\pm$ 52.6	168.1	166.7 $\pm$ 2.09	91.3	91.2 $\pm$ 0.9	200.2	231.0 $\pm$ 45.9
	621.0		524.4		649.0		165.2		92.0		212.0	
									90.3		280.7	
PS	638.9	714.9 $\pm$ 107.6	861.7	759.8 $\pm$ 144.1	654.8	744.7 $\pm$ 127.2	202.8	182.2 $\pm$ 29.1	97.2	89.9 $\pm$ 10.3	186.8	219.0 $\pm$ 68.1
	791.0		657.9		834.7		161.7		82.6		251.7	
PVP	577.0	558.7 $\pm$ 293.7	758.5	722.4 $\pm$ 430.7	723.4	771.6 $\pm$ 133.2	202.5	205.2 $\pm$ 15.7	106.0	102.3 $\pm$ 12.5	226.8	283.4 $\pm$ 43.5
	842.8		1133.9		669.2		222.1		88.3		264.4	
	256.3		274.7		922.1		191.0		112.5		359.0	

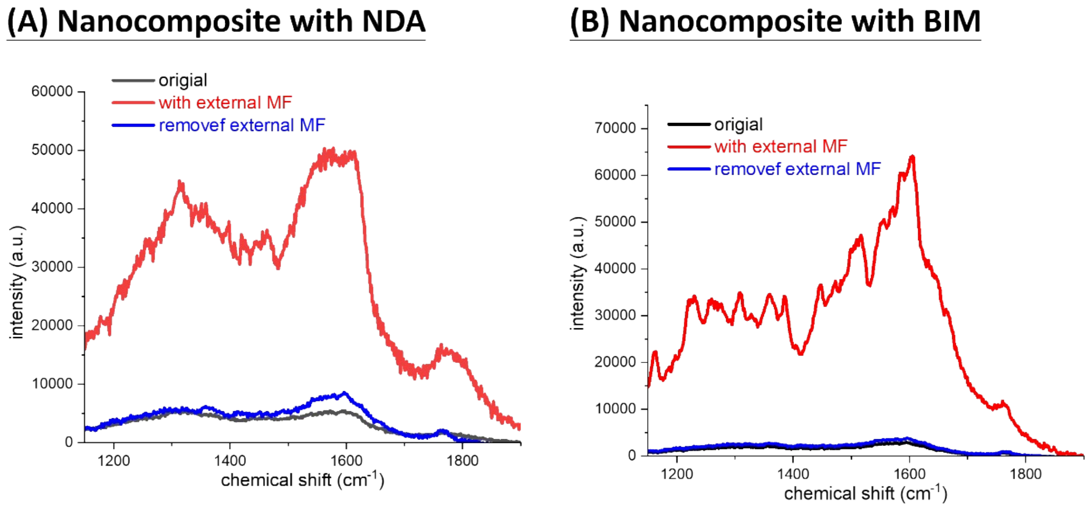


Figure S19 The reproducibility test of nanocomposites with magnetic materials for magnetic sensing application. Through the change of Raman scattering spectrum changes with the change of external MF, it can be observed that after the external MF is removed, the rearrange of molecules around the dimer nanojunction returns to the original state. This allows these nanocomposites to be reused.