

Frequency Response of MAPbI₃ Perovskites for Photodetection Application

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The frequency response of the photocurrent (I_{ph}) generated in perovskite photodetectors is a crucial issue in imaging or telecommunication applications, although it is briefly discussed in the literature. The present work obtains for the first time the complete expression of the I_{ph} generated on MAPbI₃ (MA:methylammoniumI) perovskite polycrystalline thin films. A conditioning circuit is used to extract I_{ph} at 1 V of bias voltage under a square-modulated laser excitation with a sensitivity smaller than 1 nW and a linear dynamic range LDR > 200 dB; It allows an accurate determination of the module of I_{ph} together with the phase, which is not usually reported in photodetector systems. The frequency domain analysis reveals that I_{ph} can be modeled by two fractional poles located at low (10 kHz) and high (39–250 kHz) cut-off frequencies. Optimum geometrical parameters and excitation fluence are found for the wider response, resulting for the best device on a cut off frequency up to 250 kHz, and the reproduction of square-modulated optical waves up to 100 kHz. These results represent an important strategy toward the electrical analysis of MAPbI₃ (or other perovskite materials) for the design of posterior electronic stages, optimization of devices and determination of their figures of merit.

cost-effective alternative to the traditional semiconductors to implement photoconductors, photodiodes, or phototransistors.^[3,4] For example, MAPbI₃ thin films exhibit ideal properties for photodetection, such as high absorption coefficients above the bandgap and sharp band edges,^[5] carrier diffusion lengths longer than 10 μm,^[6] and tolerance against crystalline defects.^[7] Indeed, an active MAPbI₃ layer provides similar responsivity or dynamic range with much thinner films, compared with traditional semiconductors. Moreover, they can be processed from solution, which enables a hybrid integration with other photonic or electronic systems within in a potentially simple and low-cost manner.^[8] MAPbX₃-based photodetectors are often characterized in terms of their responsivity (R), spectral tunability, high linear dynamic range, fast response time, and low noise.^[9–23]

1. Introduction

MAPbX₃ (MA:methylammonium, X = Br, Cl, and I) polycrystalline thin films have attracted significant attention due to their outstanding optoelectronic properties together with the straightforward and low-cost fabrication processes.^[1,2] Among other applications, MAPbX₃-based devices present extraordinarily high photoconversion efficiencies, which make them a promising and

However, the determination of the frequency (f) response of perovskite photodetectors is practically absent in the literature, despite being an important parameter that provides the bandwidth or the upper limit to the speed of the input signal, crucial figures of merit in image processing, data acquisition or telecommunication applications.^[12] There are only a few publications where the –3 dB cut-off frequency (f_{-3dB}) is indicated,^[9,12–14] but none of them extracts a useful expression to predict the

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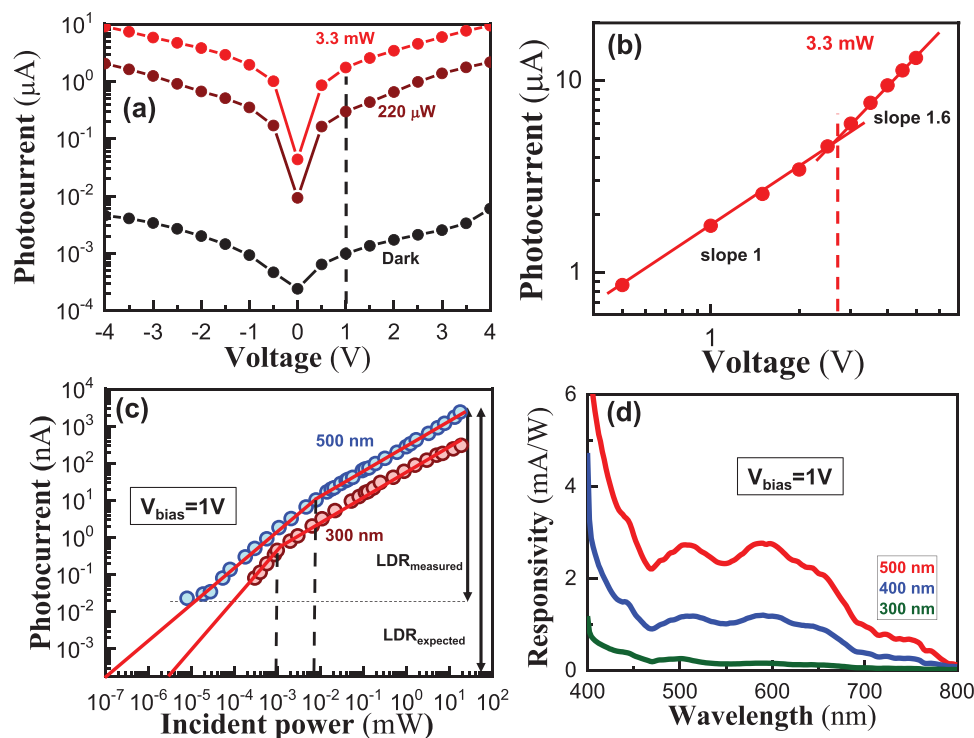


Figure 1. a) I_{ph} - V curve under different light intensities for a sample with $d = 500$ nm using an excitation laser at 450 nm. b) Log-log plot of the I - V curve ($V > 0$) for $P = 3.3$ mW. c) Log-log plot of the I_{ph} - P curve for photoconductors with MAPbI₃ thicknesses of 500 nm (blue symbols) and 300 nm (brown symbols) using the same laser at 450 nm. d) Spectral responsivity for photoconductors with MAPbI₃ thicknesses of 500 nm (red), 400 nm (blue) and 300 nm (green).

behavior of the photodevice with f . Nevertheless, its optimization or the design of additional electronic stages composing the full receptor system requires an expression in the complex plane for the generated photocurrent (I_{ph}) versus f , i.e., $I_{ph}(j\omega)/P_{in}(j\omega)$, where j is the imaginary number and Ω the angular pulsation ($\omega = 2\pi f$). For this purpose, it is mandatory to obtain experimentally the module of I_{ph} together with the phase-shift ($\Delta\phi$), which, is not usually reported because its measurement is not directly obtained with conventional electronic systems. Besides, a rigorous estimation of the transfer function requires isolating the influence of the measurement system, which, again, needs the design of an adequate conditioning circuit.

In this work, we determine the photodetection performances of MAPbI₃ polycrystalline thin films working in photoconductive mode. The device consists of the perovskite deposited on interdigital electrodes and connected to an appropriate conditioning circuit that supplies the required bias voltage, extracts the generated photocurrent with high sensitivity and filters out the dark current or other undesired noise. At these conditions, the full detector system illuminated with 450 nm light exhibits a high on-off ratio, sensitivity below 1 nW, and photocurrents smaller than 1 nA. Moreover, a frequency modulated spectroscopy technique^[24] is successfully applied to obtain the transfer function in the complex plane of I_{ph} generated in the MAPbI₃ film, extract the rise/fall times, and the cut-off frequency (80 kHz for the best device). More interestingly, an equivalent circuit model is obtained from this transfer function, which is useful for the future optimization of photodevices or the design of ad-

ditional electronic stages. This approach is applied to obtain the dependence of the transfer function with the geometrical parameters in several sample configurations and can be extrapolated to other perovskite materials. Thus, these results represent an important step toward understanding the capabilities of MAPbI₃ thin films for visible light photodetection and its potential applications in imaging, telecommunications and sensors.

2. Characterization of the Photoconductor

The photodetectors used in this work consisted of a MAPbI₃ thin film deposited on a commercial substrate with pre-patterned electrodes, see Figure S1 (Supporting Information) and Experimental Section. The films are formed by compact polycrystalline grains and present good homogeneity, see SEM image in Figure S2 (Supporting Information), and exhibit the ABX₃ crystal structure characteristic of perovskites, see in XRD Figure S3 (Supporting Information). Several sample series were prepared with a fixed electrode gap of $g = 50$ μm and a MAPbI₃ film thickness of $d = 300$, 400 and 500 nm. As previously reported with MAPbI₃ thin films,^[5,15] the absorption spectra of the film (see Figure S4, Supporting Information) present a high absorption coefficient greater than $12\ \mu\text{m}^{-1}$ at excitation wavelengths shorter than 550 nm. Figure 1a presents the I - V curve characterized under dark and continuous wave (CW) illumination of 450 nm for $d = 500$ nm (the other thicknesses are included in the Figure S5, Supporting Information). The I - V curve measured under dark conditions, black symbols in Figure 1a, indicates a dark current,

I_{dark} , smaller than 10 nA and a resistivity between 1800 and 3000 Ω cm, which are similar to values previously published for the same material.^[12,15,17,22] The films under CW illumination of 450 nm showed a remarkable photocurrent (I_{ph}), $I_{\text{ph}} \approx 1\text{--}10$ μA , even for low applied bias voltage (<3 V). The carrier mobility calculated with the Mott–Gurney law reaches $\approx 10^{-2}$ and ≈ 1 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ for dark conditions and $P = 3.3$ mW, respectively, which, again, are in agreement with the values reported for MAPbI₃ films elsewhere.^[9,16,25] The dark current and photocurrents show a negligible hysteresis when the voltage is sequentially swept from -5 to 5 V and 5 to -5 V, see Figure S5 (Supporting Information). Only the thinner layer ($d = 300$ nm) exhibits a slight difference in the cyclic I – V curves, which can be ascribed to the lower quality due to thickness in the range of the grain size. The log-log plot of the I – V curve at $P = 3.3$ mW, Figure 1b, suggests that the device behaves as a linear ohmic resistance below ≈ 3 V. This limit corresponds to the required voltage, V_{TFL} , to fill the trap states of the perovskite with the incident carriers and allows us to estimate the density of trap states, n_{trap} , by the equation:^[26]

$$n_{\text{trap}} = \frac{2 \times \epsilon_0 \times \epsilon_r \times V_{\text{TFL}}}{e \times g^2} \quad (1)$$

where ϵ_0 is the vacuum permittivity, ϵ_r the MAPbI₃ relative dielectric constant, g the channel length and e the elementary charge. Considering $\epsilon_r = 32$ reported for MAPbI₃,^[27] Equation (1) yields $n_{\text{trap}} = 4 \times 10^{12} \text{ cm}^{-3}$, which is close to the values reported for perovskite thin films measured with microgaps,^[28,29] indicating the good crystallinity and homogeneity of the films under study. Thus, to ensure that the device works in the ohmic linear regime, a bias of 1 V is fixed in the following experiments. At the conditions $V_{\text{bias}} = 1$ V, I_{ph} reaches 0.3 and 1.8 μA for $P = 220$ μW and $P = 3.3$ mW, respectively. These values correspond to a linear dynamic range ($\text{LDR} = 20 \times \log(I_{\text{ph}}/I_{\text{dark}})$) of $\text{LDR} = 49\text{--}65$ dB and a responsivity of $R = 0.5\text{--}1$ mA W^{-1} . Nevertheless, the figures of merit of the detector can be further improved with an appropriate electronic circuitry that converts I_{ph} into an accurate signal that can be processed at the downstream stages of the receptor system. With this objective, we propose the use of a transimpedance amplifier, see Figure S6 (Supporting Information), designed to set the operating point to the photoconductor, reduce the noise and discriminate low photocurrent levels on broad bandwidth. The electrical characterization of this circuit indicates an almost constant transimpedance response between direct current (DC) and 775 kHz measured for 1 V of bias voltage under modulated laser excitation at 450 nm, see Figure S7 (Supporting Information), with the required sensitivity to characterize our MAPbI₃ photoconductive devices. For this purpose, the electronic components of the circuit were properly optimized to accurately measure the bandwidth of the perovskite photodetector (see next section) and to reduce its dark current. This conditioning circuit was implemented on a printed circuit board (PCB), see Figure S8 (Supporting Information), that converts I_{ph} into a voltage that can be measured either on an oscilloscope or a lock-in amplifier, see Figure S9 (Supporting Information) and experimental Section. The PCB is particularly useful to analyze the photodetection properties under the square modulation of the incident optical beam. In that case, the influ-

ence of I_{dark} is prevented, and the overall noise is significantly reduced with a lock-in amplifier. For example, for a modulated signal of 1 kHz and $V_{\text{bias}} = 1$ V, the full detector system (photoconductor + PCB + lock-in) allows an accurate characterization of I_{ph} between $P = 8$ nW (1.4 $\mu\text{W cm}^{-2}$) and $P = 19$ mW (maximum laser fluence, 3.3 W cm^{-2}), see Figure 1c, which resulted on an increased experimental LDR = 73 dB and LDR = 100.7 dB for $d = 300$ and 500 nm, respectively. Higher LDR can be obtained by extrapolating the measured I_{ph} on the $I_{\text{ph}}\text{--}P$ curve^[12] in Figure 1c, reaching 202 dB for the best device ($d = 500$ nm), which improves the values reported with many MAPbI₃ photodetectors (see Table S1, Supporting Information). More interestingly, the $I_{\text{ph}}\text{--}P$ curve also indicates an extremely low noise equivalent power, NEP = 0.1 nW (17.5 nW cm^{-2}) for $d = 500$ nm. At these conditions, the experimental detectivity reaches 3.3×10^{11} Jones. Besides, the $I_{\text{ph}}\text{--}P$ curve presented in Figure 1c evolves from a linear ($I_{\text{ph}} \propto P$) to a sublinear ($I_{\text{ph}} \propto P^{0.7}$) slope above a threshold power of $P = 7.8$ μW (1.3 mW cm^{-2}) and $P = 1$ μW (175 $\mu\text{W cm}^{-2}$) for $d = 500$ and 300 nm, respectively, see dashed lines in the figure. This onset corresponds to the transition where the bimolecular recombination becomes dominant over the low carrier injection.^[13] Finally, the devices presented the spectral response of the responsivity (R) plotted in Figure 1d, whose shape is similar to that reported for MAPbI₃ photodetectors. Again, the photoconductors fabricated with the thicker MAPbI₃ films ($d = 500$ nm) exhibited a better response, with $R > 3$ mA W^{-1} at 450 nm and an increased R for low excitations (see Figure S10, Supporting Information). Here, the responsivity is reduced to 1–6 mA W^{-1} because of the low bias voltage applied to the electrodes, limited in our study to 1 V to avoid heating or other non-desirable effects.

3. Frequency Response

The electronic interface proposed in the present work is particularly beneficial in the amplification of I_{ph} 's generated by modulated optical beams, commonly applied in imaging, telecommunication or multiplexing applications.^[30,31] As discussed in the previous section, the circuit provides an accurate voltage proportional to I_{ph} and with an almost flat response up to 775 kHz. At these conditions, the frequency response of the I_{ph} can be obtained by measuring the circuit voltage with a lock-in amplifier synchronized with the excitation, as depicted in Figure S9 (Supporting Information). In particular, the lock-in amplifier extracts both the amplitude and the phase-shift, $\Delta\phi$, from the reference of the detected signal, which is required to obtain the transfer function of the system. Figure 2 presents the frequency response in amplitude (brown symbols) and phase-shift ($\Delta\phi$, blue symbols) experimentally obtained for three photoconductive devices with $d = 500$ nm (a,d), 400 nm (b,e) and 300 nm (c,f) illuminated at 3.3 mW (a–c) and 200 μW (d–f). As other photodetector devices,^[9,12–14] the amplitude of I_{ph} with f indicates that the devices behave as a low pass filter with a cut-off frequency that depends on the thickness of the MAPbI₃ and the excitation fluence. With the optimum conditions of $d = 500$ nm and $P = 3.3$ mW the device presents $f_{-3\text{dB}} = 80$ kHz, while it decreases down to $f_{-3\text{dB}} \approx 10$ kHz for the thinner films ($d = 300$ nm) and smaller excitations ($P = 220$ μW), see Table 1. Since improved crystallization of the MAPbI₃ film enhances the bandwidth,^[9] we attribute the

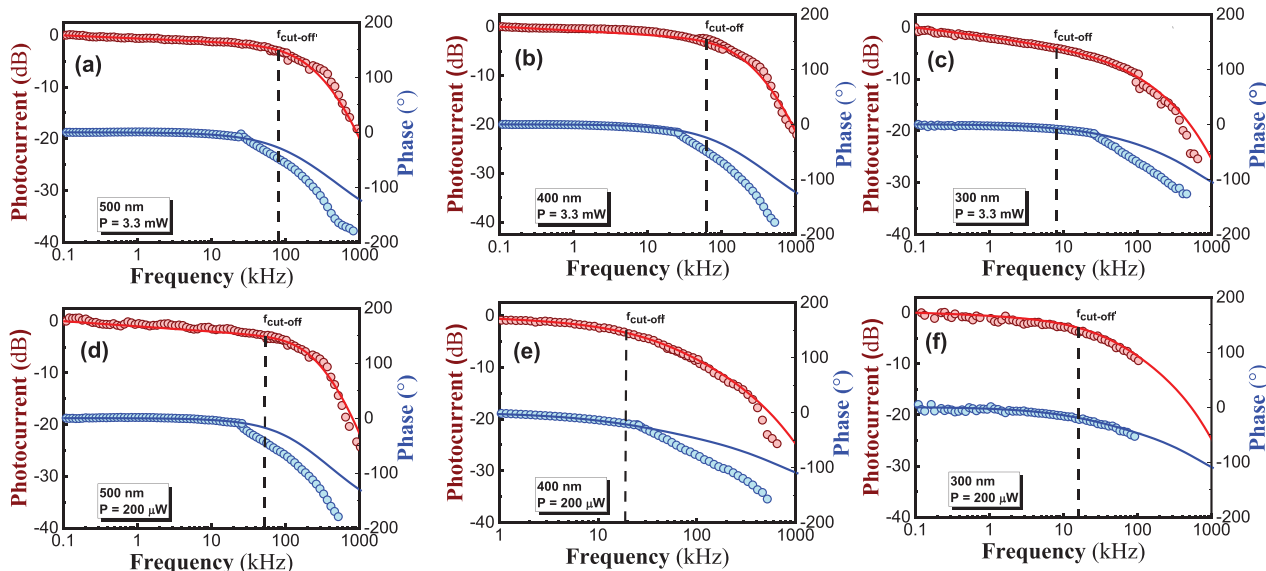


Figure 2. Frequency response of MAPbI₃ photoconductor devices under 3.3 mW (a–c) and 200 μW (d–f) and. (a,d) 500 nm, (b,e) 400 nm, (c,f) 300 nm, using a modulated laser at 450 nm. Brown and blue colors correspond to the amplitude and phase-shift of the photocurrent, respectively. Symbols and solid lines refer to the experimental points and the fitting, respectively.

superior -3 dB frequency achieved in thicker films and higher excitation fluence (80 kHz) to the improved layer formation and more intrinsic behaviour of photoconductor. It is worth noting that this value surpasses the typical -3 dB frequency with interdigital electrodes, where the bandwidth is limited to ≈ 10 kHz^[19] (refer to Table S1, Supporting Information). This limitation is close to the $f_{-3\text{ dB}}$ measured with our thinner layers (300 nm), 7.6–16.5 kHz. Only more sophisticated configurations, i.e., stacked conductive/electrode layers that reduce the RC-constant between the electrodes and the semiconductor, demonstrate broader cut-off frequencies (> 100 kHz). In the same way, $\Delta\varphi$ follows the low-pass filter response observed with the amplitude: $\Delta\varphi$ is constant for low f , and abruptly decreases above a certain f that depends on the thickness and the excitation fluence.

Moreover, the experimental amplitude/phase I_{ph} of the MAPbI₃ thin films used in this work (solid lines in Figure 2b) under different excitation ($P = 3.3$ mW, 200 μW) and geometrical conditions ($d = 500, 400,$ and 300 nm) can be simulated by a transfer function composed by three fractional poles:

$$\frac{i_{\text{ph}}}{i_{\text{ph}0}} = \frac{1}{\left(1 + \left(\frac{f}{f_{c1}}\right)^{N1}\right) \times \left(1 + \left(\frac{f}{f_{c2}}\right)^{N2}\right) \times \left(1 + \left(\frac{f}{f_{c3}}\right)^{N3}\right)} \quad (2)$$

Table 1. Spectral response as a function of the thickness and excitation fluence.

d [nm]	$f_{-3\text{ dB}}$ [kHz]	Slope [dB dec ⁻¹]	$f_{-3\text{ dB}}$ [kHz]	Slope [dB dec ⁻¹]
	P = 3.3 mW		P = 200 μW	
500	80	15.4	52	20.5
400	62	11.2	19.8	6.0
300	7.6	7.5	16.5	5.2

where $i_{\text{ph}0}$ is the photocurrent at DC and the fitting parameters f_{c_i} and N_i ($i = 1,2,3$) depend on the thickness of the layer (d) and the excitation fluence (P), see Table 2. Since the amplitude and phase depend on the same fitting parameters, a compromise must be found between the amplitude and phase fitting. In this context, we chose to prioritize amplitude over phase, as it plays a crucial role in determining the -3 dB bandwidth of the system. Under these conditions, our modeling accurately reproduces the experimental amplitude, accompanied by a reasonable fitting of the phase until the cut-off frequency, as illustrated by the solid lines in Figure 2. The phase of the proposed model begins to move away from the real for $f > f_{-3\text{ dB}}$. Nevertheless, this deviation from the model is not relevant since the interest of the modeling is in the window $f < f_{-3\text{ dB}}$, where properties of the material are manifested. The parameter $i_{\text{ph}0}$ increases with the excitation fluence following the $P^{0.7}$ law deduced in Figure 1c, and the sample volume (i.e., thickness of the film) until saturation above $d > 400$ nm, see Figure 3a. This saturation is attributed to the strong attenuation of the pump at 450 nm along the layer, leading to a negligible excitation above this thickness (i.e., $P(d) \propto e^{-\alpha d}$, where $\alpha = 12 \mu\text{m}^{-1}$). The three poles $(1 + (f/f_{c_i})^{N_i})$ necessary to fit the data indicate three different physical mechanisms

Table 2. Parameters of the transfer function.

P	d [nm]	$i_{\text{ph}0}$ [μA]	N_1	f_{c1} [MHz]	N_2	f_{c2} [MHz]	N_3	f_{c3} [MHz]
3.3 mW	500 nm	1	0.06	0.01	0.9	0.25	1	0.775
	400 nm	0.92	0.06	0.01	0.9	0.18	1	0.775
	300 nm	0.08	0.28	0.01	0.9	0.18	1	0.775
200 μW	500 nm	0.17	0.1	0.01	0.99	0.25	1	0.775
	400 nm	0.19	0.018	0.01	0.7	0.039	1	0.775
	300 nm	0.012	0.018	0.01	0.7	0.039	1	0.775

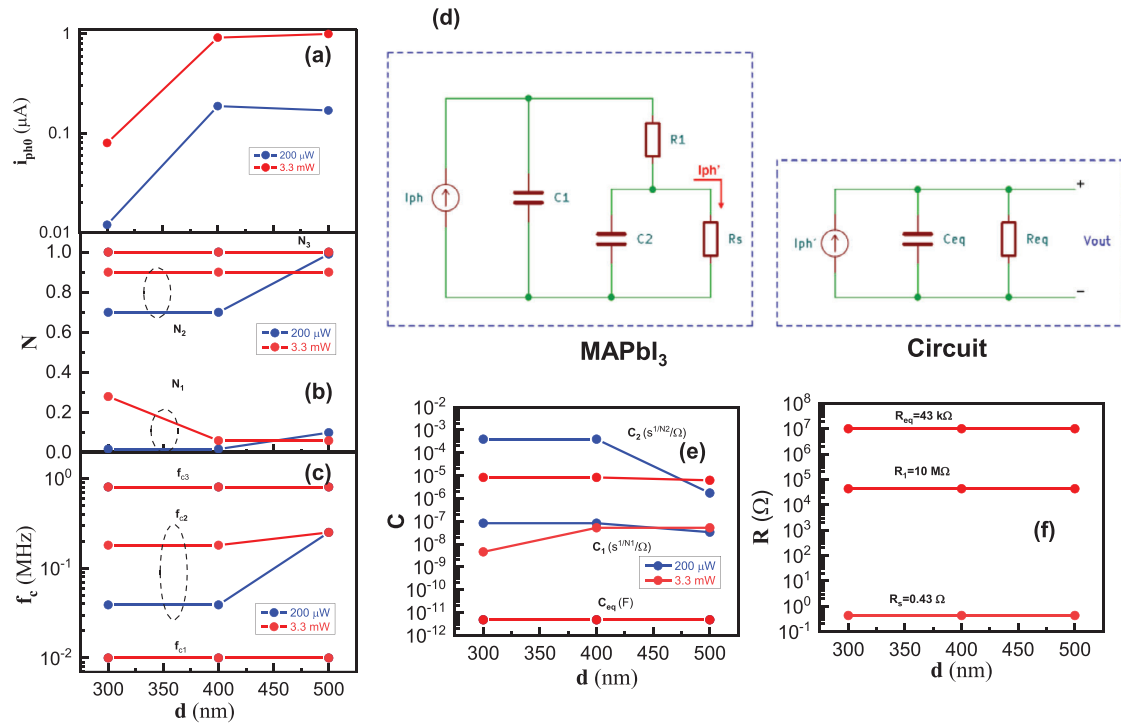


Figure 3. Parameters of the Equation (2) used to fit the data: a) i_{ph0} b) N_1, N_2, N_3 c) f_{c1}, f_{c2}, f_{c3} d) Equivalent circuit deduced from the transfer function. e) R_1, R_s, R_{eq} . f) C_1, C_2, C_{eq} . Blue and red colors correspond to an excitation of 200 and 3.3 mW, respectively.

influencing the frequency response. Figure 3b,c shows the dependence of N_i and f_{ci} on P and d . There is a low frequency pole fixed at $f_{c1} = 10$ kHz independently of P and d . The first pole is associated to the non-flat response of I_{ph} below the cut-off frequency, also observed in previous works.^[9,12–14] This pole is modeled here with a fixed frequency at $f_{c1} = 10$ kHz, independently of P and d , and a small exponent ($N_1 = 0.1–0.01$). The second pole determines the cut-off frequency of the material under study, and it is associated to the intrinsic capacity of the device. The best fitting for this pole is obtained for exponent close to one ($N_2 = 0.7–0.99$) and a frequency, f_{c2} that increases with P and d from 39 to 250 kHz, see Table II. Finally, the third pole is fixed at $f_{c1} = 775$ kHz and is ascribed to the RC constant of the conditioning circuit.

The transfer function given by Equation (2) can be synthesized by the equivalent circuit depicted in Figure 3d. For this purpose, it is useful to introduce two stages to separate the contributions of the material under study and the conditioning circuit. The MAPbI₃ is modeled by a current source dependent on the excitation fluence (P) and two R-C networks that determine the two pole frequencies. Besides, the conditioning circuit is fixed as the feedback network that sets the transimpedance gain ($R_{eq} = 43$ k Ω and $C_{eq} = 4.3$ pF). Figure 3e,f plots each value of R and C obtained from the model as function of the parameters, P , and d . The first RC network is chosen to reproduce the behavior at low frequency (DC–10 kHz). At these conditions, R_1 is fixed as the resistance of MAPbI₃ measured under CW illumination, which is reported to be 1–10 M Ω ,^[15] and fixed here to $R_1 = 10$ M Ω according to the I – V curve presented in Figure 2. Then, C_1 is calculated as the required capacitance to obtain the pole frequency,

f_{c1} . For this purpose, the particular shape of the pole requires to introduce a fractional capacitor, i.e., a capacitor with impedance $Z = 1/((j \omega)^N C)$, commonly used to model the electrical response of not pure electrical systems (vegetables, minerals...)^[32] In the MAPbI₃ films under study, $C_1 = 10^{-8}$ – 10^{-7} s ^{N_1} Ω^{-1} , with $N_1 = 0.01–0.1$, as deduced in Figure 2.^[32] Since N_1 is close to 0, the first RC network exhibits quasi-resistive behavior, wherein the deviations from a pure resistance reproduce decrease observed in the magnitude of I_{ph} for frequencies below 10 kHz, see Figure 3. Thus, C_1 can be linked to the slow response in photocurrent introduced by shallow traps.^[33] On the other hand, the second RC network is introduced to reproduce the behavior of I_{ph} at high frequencies and is correlated with the experimental $f_{c3,dB}$ obtained in Figure 3.^[9,31] Then, this network corresponds to the resistance and capacity of the MAPbI₃ film of the circuit in the alternating current regime. By fixing R_s as a shunt resistance $R_s = 0.43 \Omega$, C_s results $C_s = 10^{-6}$ – $4 \cdot 10^{-4}$ s ^{N_2} Ω^{-1} , with $N_2 = 0.7$ – 1 , see Figure 2. The exponent N_2 close to 1 indicates a quasi-pure capacitive behavior for this fractional capacitor, leading to slope of ≈ 20 dB per decade above the pole frequency, f_{c2} , that determines the cut-off. Considering $N_2 \approx 1$ and $C_s \approx 10^{-6}$ F obtained for $d = 500$ nm, the time constant derived from this network results on $R_s \cdot C_s = 0.43 \mu$ s, which gives on $f \approx 370$ kHz, close to the cut-off frequency deduced from the second network. The deviations from the theoretical value can be ascribed to the transient time (τ_t) of electrons along the photoconductive channel (τ) that decrease the bandwidth by the equation:^[31]

$$f_{-3dB}^{-2} = \left(\frac{3.5}{2 \times \pi \times \tau_t} \right)^{-2} + \left(\frac{1}{2 \times \pi \times R \times C} \right)^{-2} \quad (3)$$

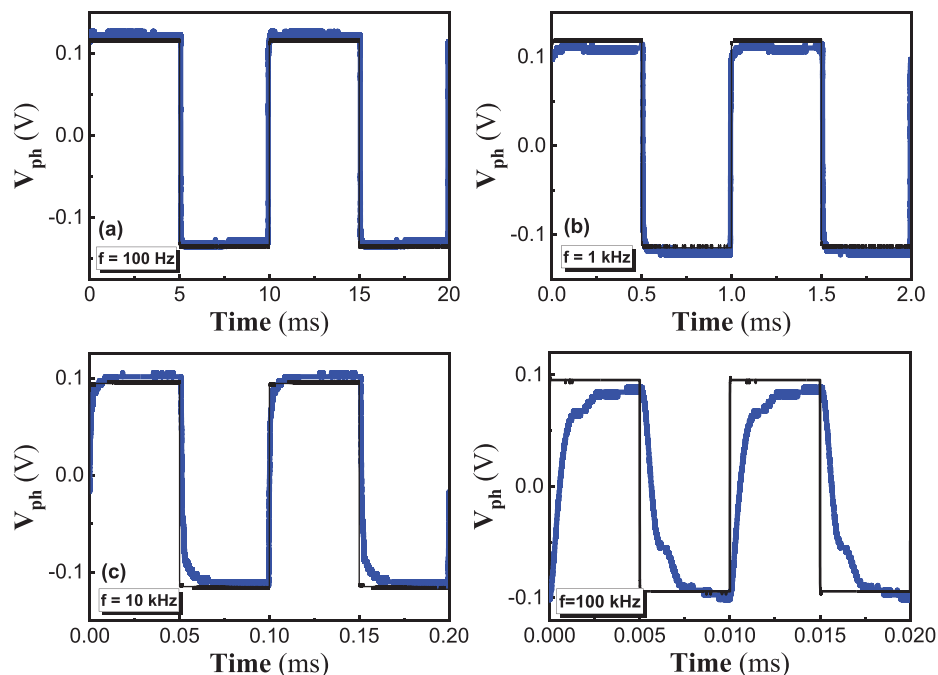


Figure 4. Detector response at a modulated wave of 3 mW (average power). a) 100 Hz, b) 1 kHz, c) 10 kHz and d) 100 kHz.

If $f_{-3\text{ dB}} = 80$ kHz, obtained experimentally for $P = 3.3$ mW and $d = 500$ nm, and $1/2 \pi RC = 270$ kHz, the transient time reaches $\tau_t = 6.6$ μs . Since our analysis indicates that the RC constant of the MAPbI₃ seen by the electrodes is the limiting factor of the frequency response. For this purpose, the strategies proposed in the literature include the improvement of crystallinity of the film^[9] or the reduction of the photodetection area.^[21,22]

Of course, the narrower bandwidths deduced with the other experimental conditions (P and d) result in longer values for τ_t . More interestingly, the highest electronic mobility (μ), as deduced through the equation $\tau_t = g^2 (\mu \text{ V})^{-1}$, is obtained for $P = 3.3$ mW and $d = 500$ nm, see Figure S7 (Supporting Information), probably because at these experimental conditions the perovskite presents a better crystallization (thicker layers) and the filling of traps (higher excitation).

The bandwidth obtained from equivalent circuit proposed in Figure 3d predicts the behavior of the device under modulated light used in imaging or telecommunications. Here, the photodetector accurately reproduces square-modulated waves up to 100 kHz, see Figure 4, and the wave is only distorted for $f > 100$ kHz, see Figure S11 (Supporting Information) for $f = 1$ MHz. Finally, the rise (t_r) and fall (t_f) times derived from the bandwidth can be estimated as $0.35/f_{-3\text{ dB}}$, where we believe that f_{c2} determined the behavior of the device rather than the $f_{-3\text{ dB}}$. Indeed, the rise/fall times obtained for $f = 100$ kHz reaches $t_r = 1.1$ μs and $t_f = 1.5$ μs , see Figure 4, which are giving $f_{-3\text{ dB}} = 230\text{--}310$ kHz, close to f_{c2} .

4. Conclusion

This manuscript analyzes the photodetection response of MAPbI₃ perovskites integrated with interdigital electrodes. A conditioning circuit is properly implemented to extract the pho-

tocurrents generated under a modulated incident excitation at 450 nm. The electronic components of this circuit were properly optimized for the case of MAPbI₃, but it can be redesigned for other materials/devices. At these conditions, we were able to detect signals smaller than 1 nW with 1 V of bias voltage with an expected LDR > 200 dB. More importantly, the analysis of generated photocurrent as a function of the frequency provided a useful expression of the complex transfer function of the material, required for the design of consecutive electronic stages, or to optimize the photodetection properties of the material. This expression reveals that the frequency response of our device is determined by two different mechanisms that can be modeled by fractional poles located at low (10 kHz) and high frequencies (39–250 kHz). The geometrical parameters (i.e., thickness of the layer) and excitation light power impact the bandwidth of the photodetector, up to 250 kHz for the optimum device. These findings contribute to the optimization of MAPbI₃ photodetectors for diverse applications where frequency response is a crucial parameter, including, but not limited, to visible communications, medical imaging, and security monitoring.

5. Experimental Section

The MAPbI₃ thin films were deposited on a commercial substrate supplied by Ossila. The commercial substrate consisted of a glass pre-patterned with interdigital ITO electrodes supplied by Ossila. The perovskite precursor solution (1.35, 1, and 0.7 M) was prepared by reacting DMF solutions (50 wt%) containing MAI, PbI₂, and DMSO (1:1:1 mol%). Then, 50 μL of this solution was spin-coated inside the glove box, at 4000 rpm (4000 ac) for 50 s. DMF was selectively washed with toluene (as antisolvent) 5–7 s after starting the spin-coating. After the deposition, a ≈ 500 nm MAPbI₃ layer covered the interdigital electrodes, see the grey area in Figure S1 (Supporting Information). The perovskite

film showed the deep brown, highly dense, shiny, and homogeneous morphology characteristic of the MAPb₃ perovskite and exhibited good stability during the measurements.

The photocurrent was measured under ambient conditions at room temperature placing the sample on an Ossila test board (P2008A1) connected to the appropriate system. The *I*–*V* curves were characterized by illuminating the samples with a continuous wave (CW) 450 nm laser with intensity ranging between 40 and 3 mW with neutral density filters and recording the photogenerated current as a function of the bias voltage using a Keithley Series 2400 Source Meter Unit (Keithley Instruments Inc). The area of the spot was 9.4×10^{-3} cm².

To characterize the frequency response, the samples were illuminated with a laser at 450 nm (MDL-XS-450, CNI laser) and its amplitude was externally modulated from 100 Hz to 2 MHz by a signal generator (TTI, model TPG3151). Here, a printed circuit board (PCB) was implemented to discriminate small signals and to analyze their evolution with the modulating frequency. The PCB was designed to provide the required bias voltage to the electrodes to convert the photocurrent *I*_{ph} generated by the MHP in an alternating voltage that can be measured by an external SR810 DSP lock-in amplifier (Stanford Research Systems) or a digital scope MSO500 (Rigol). The area of the spot was 5.6×10^{-3} cm².

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

frequency response, MAPb₃, photodetector, transimpedance amplifier

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