Phase transitions consist of sharp changes in the macroscopic properties of a physical system occurring upon the smooth variation of an external driving parameter (e.g. temperature, electric/magnetic field, . . . ). The technological applications of phase transitions are diverse, ranging from the storage of energy as latent heat [1], to the action of shape memory alloys [2]. However, phase transitions induced by radiation in the terahertz frequency range are rare. Known examples either rely on heating from the terahertz radiation to drive the phase transition indirectly [3], or require electric fields of order 1 MVcm$^{-1}$ that can only be created transiently using pulsed sources [4, 5], limiting their utility. Here we report a phase transition driven directly by a weak, continuous wave (CW) terahertz-frequency field ($\leq 1$ Vcm$^{-1}$), six orders of magnitude smaller than earlier work [4, 5].

The system described in this work consists of an atomic vapour under continuous optical excitation to an energy level with large principal quantum number $n$, a so-called Rydberg level. Driven-dissipative phase transitions occurring in such media have been a subject of recent experimental [6–8] and theoretical [9–11] interest. The phase transition induces optical bistability and has been associated with the presence of an Ising-like critical point in parameter space [12], while the bistability can be interpreted as a hysteresis cycle across an underlying first-order line [13–14]. Despite experimental progress [8] further work is needed to fully elucidate the microscopic origin of the phase transition [15] in different parameter regimes. In this work a terahertz-frequency electric field tuned close to a suitable atomic resonance perturbs the Rydberg bistability, driving the phase transition. By comparing our results with a nonlinear optical-Bloch model, we discriminate between the different mechanisms proposed as the dominating cause of the collective behaviour.

In addition to examining the underlying physical processes, we demonstrate that the abrupt change in system properties in response to a weak terahertz field makes our system suitable as a sensitive terahertz detector. Terahertz devices have seen rapid development in recent decades [16], with new technology based on media from super-conducting [17] and semi-conducting [18] solids to atomic vapour [19–21]. However the traceable calibration of terahertz detectors, typically using cryogenic bolometers, still yields substantial uncertainties [22]. Measurements of atomic properties are easy to reproduce, and therefore lend themselves naturally to measurement standards. Rydberg atoms have recently been used for absolute mm-wave intensity measurements [23, 24], using well-known atomic transition strengths as a reference. Here, by exploiting the phase transition as a terahertz detector we combine the sensitivity afforded by the discontinuous nature of the phase transition, with the ability to calibrate the device in-situ [24].

I. EXPERIMENT

Our experimental system is outlined in figure 1. The atoms of a thermal caesium vapour are continuously driven to the $21P_{3/2}$ Rydberg energy level using a three-step ladder excitation scheme [25], consisting of probe, coupling and Rydberg lasers (see methods). The vapour is monitored by photographing optical atomic
photodiode and by measuring the transmitted probe laser power, $p$, which increases when atoms are shelved in long-lived Rydberg levels or ionised [26]. We use the parameter $t = (p - p_0)/p_0$ to indicate the fractional change in transmitted laser power, where $p_0$ is the probe laser power transmitted when the Rydberg laser is far off resonance. By controlling the frequency and intensity of the Rydberg laser, the vapour can be prepared in either of two distinct steady states. The first, which we refer to as ‘Off’, is characterized by the emission of weak atomic fluorescence (figure 1b) and low probe laser transmission; the second, which we refer to as ‘On’, is characterized by the emission of weak atomic fluorescence (figure 1c). The bright orange fluorescence (figure 1d) and hold all parameters constant while the terahertz electric field amplitude is ramped up and then back down again. As the terahertz intensity increases we initially observe a quadratic rise in laser transmission (linear with intensity), before the system switches to the ‘On’ state, indicated by an abrupt increase in $t$. When the terahertz intensity is decreased again the vapour returns to the ‘Off’ state (accompanied by another sharp change in $t$), completing the full hysteresis cycle. The hysteresis cycle constitutes a strongly non-linear response of the system to the weak terahertz-frequency electric field.

To characterise the system we map the optical response to the terahertz field in the Rydberg laser- and
terahertz-detuning \{\Delta_R, \Delta_T\} plane for a selection of terahertz field amplitudes (figure 2). We describe the response by considering the regions in parameter space where the system is bistable. When the terahertz field is blocked (figure 2a), the bistability region is, as expected, independent of the terahertz field detuning. At intermediate terahertz field strength (0.15 V cm\(^{-1}\), Rydberg transition Rabi frequency \(\Omega_{\text{R}}^{\text{meas}}/2\pi = 31\) MHz) the bistable parameter space is deformed, and we see that \(\delta_\perp < 0\) for \(\Delta_T > \Delta_R\) and \(\delta_\perp > 0\) for \(\Delta_T < \Delta_R\) (figure 2b). When the field is 0.23 V cm\(^{-1}\) (\(\Omega_{\text{R}}^{\text{meas}}/2\pi = 48\) MHz) the bistable parameter space is split into two separate regions (figure 2c), and we note that bistability becomes absent for all \(\Delta_T = \Delta_R\).

II. MEAN-FIELD MODEL

A full simulation of the microscopic dynamics involved in the phase transition is computationally unfeasible, however previous work has been able to qualitatively describe the Rydberg phase transition using a mean-field model \[6,7\]. Here we use a similar model to capture the response of the system to the terahertz field (the full details of which are available in the supplementary information). For simplicity, we start from the optical-Bloch equations for a single atom, and we label \(|0\rangle\) the ground state, and \(|R\rangle\) and \(|T\rangle\) the two Rydberg energy levels, coupled by the THz field (figure 2d). As a first approximation we neglect the two intermediate states used in the experimental ladder excitation scheme, and consider a direct effective coupling between \(|0\rangle\) and \(|R\rangle\). The coherent part of the evolution is described (in a rotating-wave approximation) by the Hamiltonian

\[
\hat{H} = \left\{ \frac{\Omega_R}{2} \sigma_{0R} + \frac{\Omega_T}{2} \sigma_{RT} + \text{h.c.} \right\} - D_R\sigma_{RR} - D_{RT}\sigma_{TT},
\]

with \(\Omega_{R(T)}\) the effective Rabi frequency of the laser (terahertz field), \(D_{R(T)}\) the corresponding detuning, \(D_{RT} = D_R - D_T\), and \(\sigma_{ab} = |a\rangle \langle b|\) with \(a,b \in \{0, R, T\}\).
For the dissipative part, we consider spontaneous decay $|R\rangle \rightarrow |0\rangle (|T\rangle \rightarrow |0\rangle)$ at a rate $\Gamma_R (\Gamma_T)$. An observable $\mathcal{O}$ then evolves according to the Lindblad equation

$$\dot{\mathcal{O}} = i [\hat{H}, \mathcal{O}] + \sum_{\alpha=R,T} L_\alpha \mathcal{O} L_\alpha - \{ L_\alpha^\dagger L_\alpha, \mathcal{O} \} / 2,$$

where the jump operator $L_\alpha = \sqrt{T_\alpha} \sigma_\alpha$. In the following, we set $\Gamma_R = \Gamma_T \equiv \Gamma$.

In recent experimental work evidence that the feed-
back mechanism responsible for the bistable behaviour derives from ionised Rydberg atoms was reported [8]. The study suggests that ions created by inter-atomic collisions generate electric fields within the vapour, which in turn alter the Rydberg excitation rate through Stark shifts of the atomic energy levels. To model the effect of ionisation we assume that a fixed fraction $q_R(t)/ (1 + q_R(t))$ of the atoms in energy level $|R(T)\rangle$ spontaneously ionises, producing an ion density $n_{\text{ions}} = q_R \langle \sigma_{RR} \rangle + q_T \langle \sigma_{TT} \rangle$ of ions. We include mean-field shifts of the Rydberg levels $|R(T)\rangle$ in proportion to the ion density, which can be reabsorbed in the detunings via appropriate rescalings.

$$D_R \rightarrow D'_R = D_R - \alpha_R n_{\text{ions}},$$

$$D_{RT} \rightarrow D'_{RT} = D_{RT} - \alpha_T n_{\text{ions}},$$

where the coefficient $\alpha_{R(T)}$ is proportional to the polari-
sability of the $|R(T)\rangle$ energy level. We note that the $21S_{1/2}$ state represented by $|T\rangle$ is almost 20 times less polarisable than the $21P_{3/2}$ state represented by $|R\rangle$ [27], and so we make the approximation $\alpha_T \approx 0$.

As an “order parameter”, we focus on the density of excited atoms $N = \langle \sigma_{RR} \rangle + \langle \sigma_{TT} \rangle$, which should provide an effective qualitative comparison to the experimen-
tal data, as the transmission $t$ monotonically in-
creases with the number of atoms shelved in the Ry-
dberg energy levels [26]. The calculation results are shown in panels 2(e-g), and we note that the model reproduces several important features of the experimental data: (i) the bistability region appears at negative laser frequency detuning ($\Delta_T < 0$); (ii) the split in the bistability window occurs around the condition $\Delta_R = \Delta_T$ and; (iii) the upper bistability branch experiences a negative shift ($\delta_+ < 0$), whereas the lower one a positive shift ($\delta_- > 0$). This match was obtained via a numerical scan of the parameters, and the plots in figure 2 correspond to $\alpha_R q_R = -8.3$ and $\alpha_T q_T = -5$ in units of $\Gamma$.

The underlying mechanism leading to Rydberg bistabi-
lity has been a subject of debate. In cold atom ensem-
bles the atomic energy level shifts lead that to Rydberg “blockade” (or “anti-blockade” in the opposite case) [28]. $\Omega_{\text{D}}$ are caused by dipole interactions, and this mecha-
nism was initially invoked to explain the collective be-
aviour responsible for the vapour phase transition [5]. However, according to a recent work [15], pure van-der-
Waals interactions among excited Rydberg atoms seem to be insufficient to support bistability in a randomly distributed gas, even when thermal atomic motion pre-
vents the growth of fluctuation correlations. In the de-
velopment of the mean-field model, we trialled terms in the equations arising from resonant dipole interactions. In the mean-field picture, these pair-wise interactions are incorporated by rescaling,

$$D'_R \rightarrow D''_R = D'_R - \epsilon n_R,$$

$$D'_{RT} \rightarrow D''_{RT} = D'_{RT} - \gamma n_T,$$

where $\epsilon, \gamma$ are phenomenological parameters character-
ising the strength of the interactions. However, when these terms are dominant ($\alpha_{R(T)} \ll \epsilon, \gamma$) the simula-
tion does not match the behaviour observed in the ex-
periment. Specifically, both branches display a positive shift $\delta_+ > 0$ and, furthermore, if $\gamma$ is very large then the break in the bistability window does not occur at $\Delta_T = \Delta_R$. This suggests that dipole interactions do not dominate and instead ionisation plays the leading role in the feedback responsible for the phase transition.

III. SENSING APPLICATIONS

The edges of the hysteresis profile constitute sharp spec-
tral features which are sensitive to the presence of the ter-
ahertz radiation, and so we propose utilising the col-
lective behaviour as a fast and sensitive way to measure narrowband terahertz radiation. Furthermore the com-
plexity of the phase diagram provides the opportunity to exploit unconventional measurement protocols. In Section 1(a) we noted that cycling the terahertz intensity can result in a complete hysteresis loop, however this is not necessarily the case. If the laser frequency is set so that the system is bistable when the terahertz field has zero intensity ($\Delta_T < \Delta_R < \Delta_+$), the hysteresis loop opens and we see a latching response (figure 3a). In this case $t$ increases steadily as the terahertz intensity is ramped up, until the system undergoes the transition to the ‘On’ phase, giving a sharp increase in $t$. However, if the ter-
ahertz field intensity subsequently returns to zero, the transition back to the ‘Off’ state is absent. Instead the system remains in the ‘On’ state for as long as the control parameters do not drift, effectively latching in an altered state. To gain full control of the ‘On’ and ‘Off’ states of the vapour using the terahertz field alone, it would be necessary to stabilise Rydberg laser to a de-
tuning, $\Delta_m$, that divides the two branches of the bistable parameter space when terahertz field is at maximum inten-
sity (indicated by the dashed line in figure 2c). In this circumstance we would expect a pulse of terahertz radiation with $\Delta_T > \Delta_m$ to transfer the system from ‘Off’ to ‘On’ (as we demonstrate with the latching de-
tector configuration), and a pulse with $\Delta_T < \Delta_m$ to reverse the operation, taking ‘On’ back to ‘Off’.

The result of implementing the system as a latching de-
tector is shown in Figure 3b. After the vapour is ini-
terahertz pulse is ‘detected’ and flips the system from ‘Off’ to ‘On’. The system remains in its altered state until the system is reset by cycling the laser power. (c) Latching response time: We show the same latching response on a microsecond timescale. Although the terahertz pulse is constrained to last 1 ms we see that the vapour takes only 20 μs to respond. (d) Frequency shift of bistability boundaries: The frequency shifts follow a linear relationship with intensity in figure 3d (error bars show the standard error in the mean of repeated measurements, each lasting 1 ms).

Finally we show how to implement a detector with linear response, which - beyond speed and high sensitivity - is often a desirable property. To linearise the detector output we demonstrate a separate protocol, making use of the frequency shift of the laser detuning range for which the vapour is bistable. We repeatedly scan the Rydberg laser detuning (1 ms per cycle) and read Δ' from each scan. The shifts δ+, δ− and Σ = δ+ + δ− are then calculated using a reference measurement of Δ±. With the terahertz detuning set to ΔT/2π = −91 MHz such that ΔT > Δ−, we show the dependence of the shifts on the terahertz intensity in figure 3d (error bars show the standard error in the mean of repeated measurements). The frequency shifts follow a linear relationship with intensity, making intensity (rather than electric field amplitude) the natural sensitivity of the detector (for reference 30 mWm−2 is equivalent to 0.048 Vcm−1). By fitting straight lines constrained to pass through the origin we deduce slope coefficients, mδ±,Σ, which we combine with the average error of the data points, σ.
and the measurement time, $\tau$, to find an effective intensity NEP, $\sqrt{\sigma/m_2} = 48 \pm 3 \, \mu\text{Wm}^{-2}\text{Hz}^{-1/2}$. Taking the detector area as the probe laser beam cross section yields $\text{NEP} \leq 1 \, \text{pWHz}^{1/2}$, though it is not clear how the noise will scale with the laser beam area. Consecutive measurements at 200 nanosecond intervals show no correlation, indicating that the noise present in the system is white in character.

We reference the terahertz field amplitude by making a direct, in-situ measurement of the Rabi-driving frequency of the Rydberg transition driven by the terahertz field. Combining the Rabi frequency with knowledge of the atomic dipole matrix element $27$ then allows a calculation of the field amplitude. The measurement is performed by setting the terahertz field detuning to zero ($\Delta T = 0$) and reading out the frequency interval between a pair of spectral features in the probe laser transmission (Autler-Townes splitting $28$). The result is an absolute measurement of the electric field amplitude, which can be traced directly to fundamental units. The ‘Autler-Townes’ method has been used as a sensitive probe of microwave $29$ and terahertz $30$ fields in its own right, and techniques such as homodyne measurement $41$, frequency modulation spectroscopy $42$ and lock-in amplification $39$ have been employed to further improve the sensitivity. The results achieved by the Autler-Townes method are now limited by the spectral linewidth of the Rydberg transition and the shot noise of the probe laser $42$, which is particularly relevant because the method requires the probe laser to be in the weak excitation regime. Although implementing similar noise reduction techniques in our system will be made complicated by the hysteresis, the collective response produces spectral features (‘Off’ to ‘On’ collective transitions) much narrower than the linewidth of single-atom spectroscopic features, which are limited by the Rydberg atom lifetime. Furthermore the use of bright excitation lasers (necessary for sufficient excited atomic population to see collective behaviour) mitigates the limit imposed by laser shot noise.

**IV. CONCLUSION**

We have demonstrated a phase transition in a thermal atomic vapour driven by a weak ($\leq 1 \, \text{Vm}^{-1}$) terahertz-frequency electric field. The necessary field strength is smaller than reported in other systems by over 6 orders of magnitude $44$. The strong, non-linear response is due to both the inherent inter-particle interactions in the vapour, and the large electric dipole coupling between the Rydberg atoms and the terahertz-frequency field. Non-linear effects induced by terahertz fields have been extensively studied $33$, with applications ranging from non-linear spectroscopy $41$ and high-harmonic generation $45$, to the search for ferroelectric domain switchings $46$. Yet such demonstrations rely on high intensity pulsed terahertz sources. By working in the vicinity of a phase transition we have shown a non-linear response to terahertz radiation in the CW regime, including permanent alteration of the state of the system.

The system can be configured as a narrowband terahertz detector, already showing performance comparable to state-of-the-art room-temperature terahertz detectors $32$. Using an atomic vapour to measure terahertz fields has particular promise because Rydberg terahertz electrometry allows for absolute calibration to SI units through the well known Rydberg atomic dipole moments $24$. Although the requirement to work near an atomic resonance restricts the choice of terahertz frequencies capable of driving such a phase transition, we note that suitable alkali-metal atom Rydberg transitions span the microwave and terahertz frequency regimes, giving thorough coverage $31$. We anticipate further applications combining the phase transition with Rydberg electrometry $39$ and Rydberg-fluorescence terahertz imaging $21$.

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Laser Excitation: We use a three-step excitation process to create Rydberg atoms in the target state. The process begins with the probe laser (820 nm) exciting the 6P\(1/2\) state to the 7S\(1/2\) state. Then, the coupling laser (1470 nm) transfers the atoms to the 21P\(3/2\) state, and finally, the Rydberg laser (2153 nm) excites the atoms to the 21S\(1/2\) state. The Rydberg atoms are then detected using a transition in the optical fields.

Atomic Vapour: The atomic vapour is contained in a quartz cell with a path length of 2 mm. The temperature of the vapour is stabilised around 70 \(\pm 0.1\) \(\)K by two ovens which encase the glass cell, one constructed from stainless steel and the other from a Teflon plate.

Methods

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state to the $21S_{1/2}$ state. The terahertz beam propagates along the axis of the laser beams.

**Automated Control:** The main experimental parameters (power and detuning of the Rydberg laser and terahertz beams) are controlled from a computer using a *LabView* program. The microwave source and terahertz AMC are controlled directly through their respective interfaces, and the Rydberg laser frequency is controlled in two ways: For slow frequency scans (figures 1, 2 and 3a) the computer scans the reference etalon to which the Rydberg laser frequency is stabilised. For fast frequency scans (figure 3d) an acousto-optic modulator (AOM) is used instead, however the range is limited to $\leq 100\text{ MHz}$. The power of the Rydberg laser is controlled using the same AOM. The automated control allowed fast data collection, permitting the data shown in figure 2 to be recorded in only few minutes.

**Experimental Parameters:** The data were recorded on separate occasions, with parameters summarised as follows:

|                     | Unit | Figure 1, 3a-c | Figure 2 | Figure 3d |
|---------------------|------|----------------|----------|-----------|
| Vapour temperature  | °C   | 71             | 77       | 71        |
| Probe laser $1/e^2$ radius | mm   | 0.06           | 0.03     | 0.03      |
| Coupling laser $1/e^2$ radius | mm   | 0.05           | 0.10     | 0.10      |
| Rydberg laser $1/e^2$ radius | mm   | 0.06           | 0.13     | 0.13      |
| Probe laser power   | $\mu$W | 40            | 70       | 30        |
| Coupling laser power| $\mu$W | 60            | 30       | 140       |
| Rydberg laser power | mW   | 330           | 310      | 410       |

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