

## Supplementary information

Here we present further details of our calculations of systematic EDM limits, and describe the additional tests and consistency checks that we applied to the dataset.

### Geometric phase

We calculate a worst case example of the edm from a geometric phase. The geometric phase is equal to the solid angle subtended by the rotation of the electric field [21]. Our electric field map [16] (Method) determines how the plate spacing varies along the beam direction  $y$ , from which we know that the field rotates around the  $x$  axis (figure 1) by less than  $\pm 0.5$  mrad. From the geometry of the plates, we expect a similar field rotation around the  $y$  axis. However, these rotations do not change sense when the electric field is reversed and therefore do not generate a systematic error. To result in an apparent EDM phase they must work in concert with a rotation that does change sense when  $E$  is reversed. Such a rotation would result for example from patch fields due to local variation of the composition, crystalline structure, or contaminants on the plate surfaces. As a worst case, we imagine a 1 V rectangular patch filling the second half of the interferometer and covering half the width of one plate. In the rest frame of the molecules this sweeps out a solid angle whose sign reverses when the applied field is reversed, causing a systematic error. After averaging over the molecular beam trajectories we calculate that the false EDM in this example is  $3 \times 10^{-30} e \cdot \text{cm}$ , which we take in table 1 as an upper limit.

### Leakage currents

Let us assume that the 1 nA average leakage current somehow manages to flow up the edge of one plate and down the opposite edge of the other over half the full length of the plates. Over the path of the molecules, this generates a mean  $B_z$  field of 5 fT. We take the corresponding false EDM,  $|\mu_B B_z / E_{\text{eff}}| = 0.2 \times 10^{-28} e \cdot \text{cm}$ , as the upper limit of this effect and treat it in table 1 as an additional uncertainty.

### Other systematic checks

The 511 channels other than  $\{E \cdot B\}$  have been examined to ensure that the experiment operates correctly. Most are expected to be consistent with zero and we find that they are. A false EDM is generated if, for any combination  $X$  of the switched parameters, there are channels  $\{X \cdot B\}$  and  $\{X \cdot E\}$  that are both non-zero. Searching all channels, we find that only RF1F and RF2F have this property, and we have already corrected for these in the analysis described in the main text.

We also analyse the early- and late-arriving halves of the probe pulses separately, and find that the EDMs derived from each are consistent.

As well as the main data from the probe detector, we analyze the data from the pump detector, a magnetometer between the shields, three magnetometers placed around the lab, two leakage current monitors, a battery dummy-input and a short-circuit dummy-input (Methods). We use the same analysis routines, searching for signals that correlate with any of the switches. From all the channels of all these detectors, we find only three that are expected to be consistent with zero but are not. The magnetometer between the shields registers a small magnetic field that correlates with the state of the rf-phase switcher, showing that the field generated by this device is different in its two states. This field is too small to be of concern and in any case does not depend on  $E$ . The magnetometer that is close to the electric field relay measures a magnetic field that correlates with  $E$ , but none of the other magnetometers register this field showing that it falls off too rapidly with distance to have any significant effect on the molecules (this fact is also shown by the first line of table 1). The two leakage current monitors register a small signal correlated with  $B$ , but this is much too small to be of concern.