High-Energy Molecular Beam Source Using a Small Shock Tube: Evaluation of Convergent Type Design

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Abstract. Molecular beam source using a small shock tube has the potential to frequently generate high energy molecular beam in a range of 1 - 5 eV without any undesirable impurities. We measured shock Mach numbers in 2 and 4-mm-diameter straight tubes to know about the propagation of shock wave in a very small shock tube. In addition, we measured shock Mach numbers in convergent shock tubes of which diameters linearly decrease from 4 mm to 2 mm, which demonstrated the possibility of a convergent shock tube to generate higher energy molecular beam than straight one.

Keywords: Shock tube, Molecular beam, Compressible flow.
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INTRODUCTION

The molecular beam technique is one of the powerful tools to analyze gas-surface interactions. Various methods have been developed to generate the beam with higher translational energy than thermal beam, of which translational energy is far less than the typical activation energy of surface reactions. Although seeded beam combined with a heated nozzle has often been used, heating temperature is limited because of nozzle material, and molecular beam contains career gas as well. Arc-heated beam has the translational energy of 1 - 5 eV. The problem is, however, that the beam contains copper atoms generated by electrode erosion and thus contaminates surfaces. In order to overcome the shortcomings of the conventional beam sources for the energy range of 1 - 5 eV, we have been developing a beam source with a non-diaphragm type small shock tube [1,2], which makes it possible to save the time of replacing a diaphragm and to operate at a repetition rate high enough for efficient data acquisition. Our goal is to generate molecular beam with the translational energy of more than 1 eV at the frequency of 1 Hz. It is noteworthy that the inner diameter of our shock tube is a few millimeters, far smaller than that of conventional shock tubes. The reduction of the volume leads to the shorter evacuation time between each shot, and this leads to a very frequent generation of molecular beam. In addition, we developed a current-loop valve which separates a tube from a high pressure room. It is actuated by the magnetic repulsion induced by opposing current [1]. Our preliminary measurement of the time-of-flight (TOF) distributions of shock-heated beam demonstrated the possibility of controlling the translational energy of the beam with the initial pressure ratio of a shock tube [2].

In the present study, we have worked on the optimization of a tube geometry in order to generate higher energy molecular beam. Care must be taken in designing a small shock tube, since it is not clear whether we can apply the design criteria for conventional large shock tubes to our small shock tube. For this reason, we have worked on revealing the propagation of shock wave in a very small shock tube. First, we estimated shock Mach numbers in small tubes by means of the quasi-one dimensional calculations. Second, we measured shock Mach numbers in 2 and 4-mm-diameter straight shock tubes. The results suggest that shock wave accelerates in short distance (≈ 200 - 300 mm) and the viscosity effect has a great effect on the damping of shock wave in a small shock tube.

In addition, it has been reported that a convergent shock tube can generate the shock wave with higher Mach number than straight one [3,4]. Stronger shock wave makes the translational energy of molecular beam higher. The design criteria for small tube diameters, however, has not been obtained yet. For this reason, we measured shock Mach numbers in convergent small tubes to reveal the propagation of shock wave, and evaluated the convergent type design. Our objective in this paper is to obtain the knowledge about the optimization of the geometry of a small shock tube in order to make the translational energy of molecular beam as high as possible.

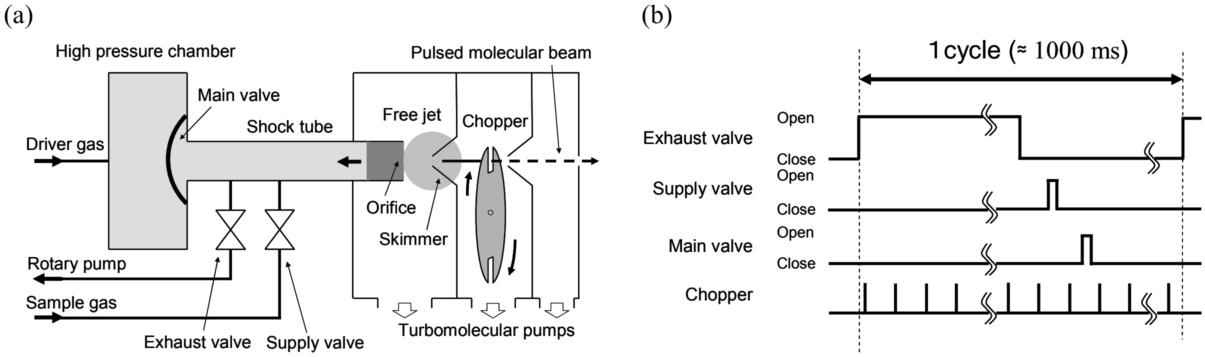

FIGURE 1. The molecular beam source with a non-diaphragm small shock tube; the schematic diagram (a) and the timing chart for the operation (b).

OVERVIEW OF THE DEDIGN

Figure 1(a) shows the schematic diagram of our molecular beam source, which consists of a non-diaphragm type shock tube and three differential pumping stages evacuated by turbomolecular pumps. High temperature gas behind reflected shock wave is extracted as a free jet through a 200-μm-diameter orifice at the tube end. The beam is collimated by the skimmer placed between the first and the second stage, and is modulated by a two-slit chopper rotating at 100 Hz with 0.4% duty cycle. Finally the diffusive component is eliminated in the third chamber before the beam enters the measurement chamber.

Several researchers also investigated shock-heated beam sources [5,6]. The replacement of diaphragm after each shot, however, makes them impractical for scattering experiments of gas molecules on surfaces, since the low signal-to-noise ratio requires signal accumulation for a large number of beam pulses. This is the reason why we have developed a high-speed valve which employs a current-loop mechanism (see Fig. 3(b)). It operates repeatedly without time-consuming replacement of the diaphragm and opens completely within about 100 μs. The shorter time to open the valve leads to the reduction of shock formation distance [7]. In addition, reducing the volume of a tube makes the evacuation time between each shot shorter, and leads to generation of molecular beam at a repetition rate high enough for efficient data acquisition.

The shock tubes used for the measurement of shock Mach numbers are as follows: straight tubes with the inner diameter of 2 and 4 mm, and convergent tubes of which diameters linearly decrease from 4 mm to 2 mm in the length of 100 mm. The length of a shock tube for a molecular beam source will be decided so that shock Mach number reaches its maximum value at the tube end.

Figure 1(b) shows the timing chart for the operation of each valve. First the residual gas in the shock tube is evacuated through the exhaust valve. The sample gas is then filled up to the target pressure through the supply valve. Finally the main valve opens synchronously with the chopper rotation so that the chopper slits pass the beam axis when the shock-heated free jet is generated. The translational energy of the beam is controlled by the initial pressure ratio of the driver gas to the sample gas in the shock tube.

NUMERICAL ANALYSIS

In order to figure out the propagation of shock wave in very small tubes, we estimated shock Mach numbers by means of quasi-one dimensional calculations. The governing equations were the conservation of mass, momentum, and energy. Addition of the equation of state made a system closed. The valve opening time (100 μs) was simulated by changing the flow passage area at the position of the valve as a function of time. The wall shear stress is given by

$$\tau_w = C_f \frac{1}{2} \rho u^2 , \tag{1}$$

where C_f represents the coefficient of pipe friction, ρ the density, and u the velocity. The coefficient of pipe friction C_f depends on the flow field. For simplicity, we set $C_f = 0.004$ in these calculations (It has been reported that C_f is

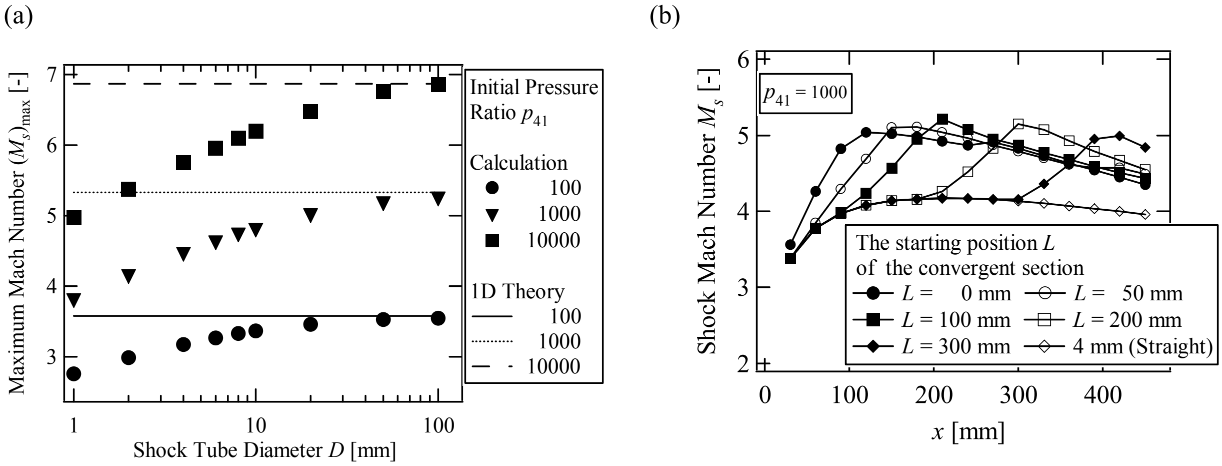

FIGURE 2. The results of numerical calculations. (a) Maximum Mach numbers $(M_s)_{\max}$ as a function of the shock tube diameter D. (b) Shock Mach numbers M_s in convergent tubes of which diameters linearly decrease from 4 mm to 2 mm in the section of $L \leq x \leq L + 100$ mm (L = 0, 50, 100, 200, and 300 mm).

about 0.003 - 0.004 in supersonic flows [8]). The governing equations were integrated using the 2nd-order AUSM-DV [9] in space and the 2nd-order Runge-Kutta in time.

In general, shock Mach number increases until it reaches its maximum value, and then decreases. The acceleration process of shock wave is attributed to the valve opening time. In other words, compression waves from the contact surface between the driver and driven gas overtake the shock wave and strengthen the wave front. On the other hand, the attenuation process of shock wave is attributed to wall boundary layer. Specifically, expansion waves from the wall boundary layer overtake the shock wave and weaken the wave front.

Figure 2(a) shows maximum Mach numbers $(M_s)_{\max}$ at the initial pressure ratio p_{41} = 100, 1000 and 10000 as a function of the shock tube diameter D, and also the theoretical line based on the one dimensional theory. For any initial pressure ratio, the maximum Mach number decreases for the smaller tube diameter since the effect of wall boundary layer becomes dominant in the smaller shock tube. This indicates that the tube diameter should be larger in order to generate higher energy shock-heated beam. On the other hand, the larger diameter leads to the larger volume of a tube, which makes the evacuation time between each shot longer. Therefore, the trade-off between shock wave attenuation and evacuation time should be taken into account in deciding tube diameter.

Figure 2(b) shows shock Mach numbers M_s at the initial pressure ratio p_{41} = 1000 in the five different geometries of convergent tubes as a function of the distance x from the valve. The diameters of each shock tube linearly decrease from 4 mm to 2 mm in the length of 100 mm, and the starting positions L of the convergent section are 0, 50, 100, 200, and 300 mm respectively (see Fig. 3(a)). The shock Mach number in a 4-mm-diameter straight tube is also shown in Fig. 2(b). The shock Mach number sharply increases in the convergent section and reaches its maximum value, which is higher than that in the straight tube, and gradually decreases afterwards. This result indicates that a convergent shock tube even with very small diameter can generate stronger shock wave than straight one. Therefore, by optimizing a tube geometry to make shock Mach number maximum at the tube end, it is expected that a convergent shock tube could generate the beam with higher translational energy than straight one.

METHOD OF EXPERIMENTS

First we measured shock Mach numbers in two straight shock tubes. Second we measured shock Mach numbers in convergent shock tubes, and evaluated the convergent type design.

The dimensions of two straight tubes are 2 and 4 mm in diameter and 500 mm in length. The geometry of a convergent shock tube is shown in Fig. 3(a). The diameters of the convergent tubes linearly decrease from 4 mm to 2 mm in the length of 100 mm, and the starting positions L of the convergent section are 50, 100, 200, and 385 mm.

The main valve, which separates the high pressure room from the low pressure room, exploits current-loop mechanisms [10] and opens completely within about 100 μs [1,2]. The schematic diagram of the valve is shown in

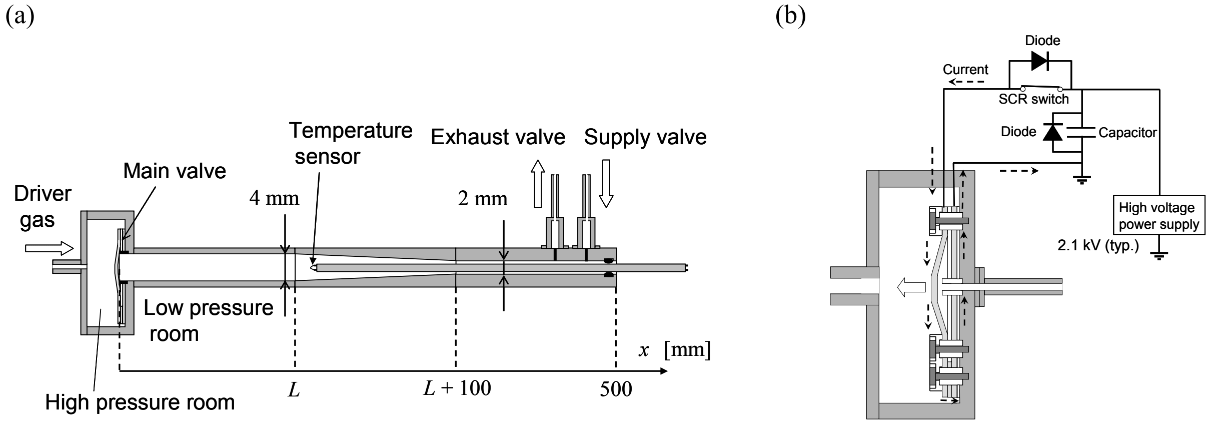

FIGURE 3. The schematic diagram of the experimental apparatus. (a) The convergent tube of which diameter linearly decreases in the section of $L \leq x \leq L + 100$ mm. The length of the tube is 500 mm. The temperature sensor is inserted from the tube end, and can be placed at arbitrary position x. (b) The current-loop valve which opens completely within about 100 μs.

Fig. 3(b). The metal plate seals the tube by resting on the elastomer tube. The sealing plate is pushed away by magnetic repulsion induced by opposing pulse current with the width of about 10 μs, and then the driver gas flows into the tube.

Commercial magnetic valves (Parker-Hanefin 009-400/279-900) are adopted for the exhaust and supply valves. Helium at 1 MPa ($\equiv p_4$) is used for the driver gas. Nitrogen is used for the driven gas, and its pressure p_1 is controlled in a range between 2×10^2 - 1×10^4 Pa. Hence the initial pressure ratio p_{41} ($= p_4 / p_1$) is 100 - 5000.

We measured the arrival time of a shock front using the high-speed response temperature sensor with a response time of less than 5 μs [2]. The thin platinum wire (diameter: 5 μm) on the front of the sensor is heated by high-temperature gas behind shock wave. The resistance of the wire then rapidly changes. The resistance change is measured with a bridge circuit. The sensor is inserted from the tube end, and can be moved along the tube axis so that the arrival time t can be measured at arbitrary position x (see Fig. 3(a)). We can make x - t diagrams by repeatedly measuring the arrival time t at the position x at the same initial pressure ratio, since the main valve opens in a reproducible fashion. Shock Mach number is determined from the slope of the line fitted by least-squares approximation to the x - t diagram.

RESULTS AND DISCUSSION

Figure 4(a) shows shock Mach numbers M_s at the initial pressure ratio p_{41} = 2000 in the 2 and 4-mm-diameter straight tubes as a function of the distance x from the main valve. The temperature T_5 of the sample gas behind reflected shock wave can be calculated as a function of the shock Mach number. Assuming that the free jet of the shock-heated gas expands substantially until the temperature becomes significantly lower than the stagnation temperature T_5, the translational energy of molecular beam is approximately given by

$$\langle E \rangle \approx \frac{\gamma_4}{\gamma_4 - 1} k_B T_5, \tag{2}$$

where γ_4 represents the ratio of specific heat of the sample gas and k_B the Boltzmann constant. The vertical axis on the right side in Fig. 4(a) shows the translational energy of the beam estimated from Eq. (2). Regardless of tube diameter, shock wave accelerates until shock Mach number reaches its maximum value around $x \approx$ 200 - 300 mm, and then attenuates. The shorter opening time of the valve makes it possible to reduce shock formation distance compared to that of conventional shock tubes. The larger diameter tube, in which the viscosity effect is less dominant, exhibits higher shock Mach numbers at all measurement points. The difference between two tubes becomes significant in the attenuation process, where shock Mach number decreases more rapidly in the smaller diameter tube.

Figure 4(b) shows maximum Mach numbers $(M_s)_{\max}$ in the straight tubes as a function of the initial pressure ratio p_{41}, and also the theoretical curve based on the one dimensional theory. The larger initial pressure ratio leads to the larger maximum Mach number. The maximum Mach number in the 4-mm-diameter tube is larger than that in the 2-mm-diameter tube for any initial pressure ratio, since the effect of wall boundary layer is less dominant in the larger diameter tube. In addition, as the initial pressure ratio increases, the deviation of the maximum Mach number from the theoretical curve becomes larger. It is because the larger initial pressure ratio leads to the faster shock velocity, and thus makes the wall shear stress larger. This figure indicates that the 4-mm-diameter tube can generate molecular beam with the translational energy of about 1 eV at $p_{41} \approx 1000$. It should be, however, noted again that the evacuation time between each shot is longer in the larger diameter tube.

Figure 5(a) shows shock Mach numbers M_s in the convergent tubes with three different geometries (L= 50, 200, and 385 mm) at $p_{41} = 1000$ as a function of the distance x from the main valve. The length of the convergent section is 100 mm for all the cases. The shock Mach number in the 4-mm-diameter straight tube is also shown in Fig. 5(a). Shock Mach number in any geometry of convergent tubes sharply increases in the convergent section and reaches its maximum value at the end of the convergent section, and then decreases because of the viscosity effect. This result suggests that it would be the best to extract a free jet at the end of the convergent section in order to make the translational energy of the beam as high as possible.

Although the shock Mach number sharply increases in the convergent section regardless of the starting positions L, maximum values $(M_s)_{\max}$ are different for each geometry. Converging the tube shape at the initial stage of the acceleration process (i.e., L = 50 mm) or at the attenuation process (i.e., L = 385 mm) leads to the lower maximum Mach number. These results indicate that the tube converging at the final stage of the acceleration process (i.e., L = 200 mm) would be the best to obtain the highest maximum Mach number.

Figure 5(b) shows maximum Mach numbers $(M_s)_{\max}$ in the 4-mm-diameter straight tube and the convergent tube of which diameter linearly decreases from 4 mm to 2 mm in the section of $100 \leq x \leq 200$ mm (L = 100 mm) as a function of the initial pressure ratio p_{41}. The theoretical curve [3] is also plotted in Fig. 5(b). For any initial pressure ratio, the maximum Mach number in the convergent tube is higher than that in the straight tube. This result indicates that a convergent shock tube could generate molecular beam with higher energy than straight one by optimizing the tube geometry. The convergent shock tube used in our experiments would generate the molecular beam with the translational energy of nearly 2 eV at $p_{41} = 5000$. It should be, however, considered that the higher initial pressure ratio leads to the lower pressure of the sample gas, which makes the intensity of molecular beam weaker.

CONCLUSION

We have investigated the influence of a tube geometry on the propagation of shock wave in order to develop a molecular beam source using a small shock tube. The quasi-one dimensional calculations indicate that wall boundary layer has a significant effect on the Mach number of shock wave propagating in the tubes with the inner diameter of a few millimeters. We measured shock Mach numbers in 2 and 4-mm-diamter straight shock tubes and

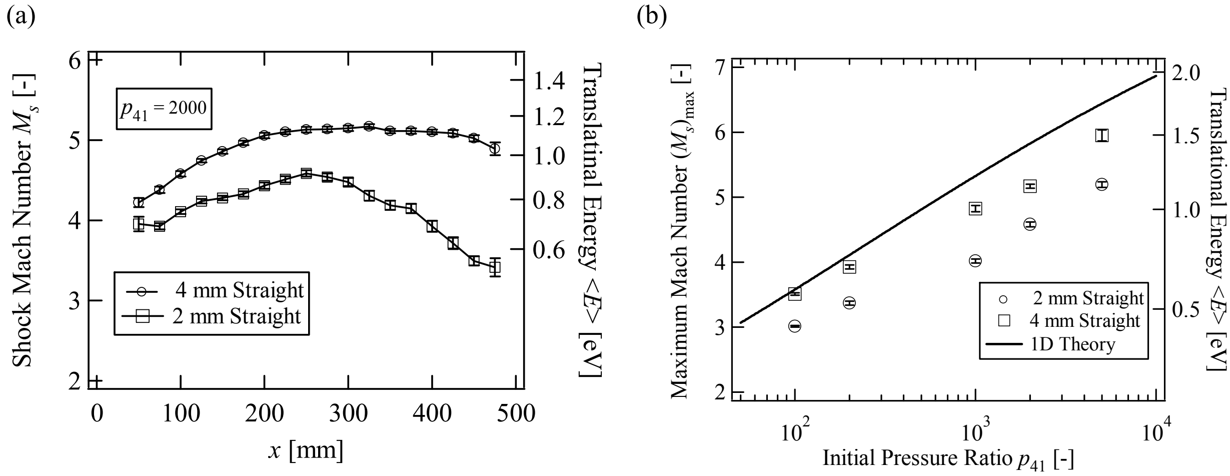

FIGURE 4. Shock Mach numbers in 2 and 4-mm-diameter straight shock tubes. (a) Shock Mach numbers M_s at the initial pressure ratio $p_{41} = 2000$ as a function of the distance x from the main valve. (b) Maximum Mach numbers $(M_s)_{\max}$ as a function of the initial pressure ratio p_{41}.

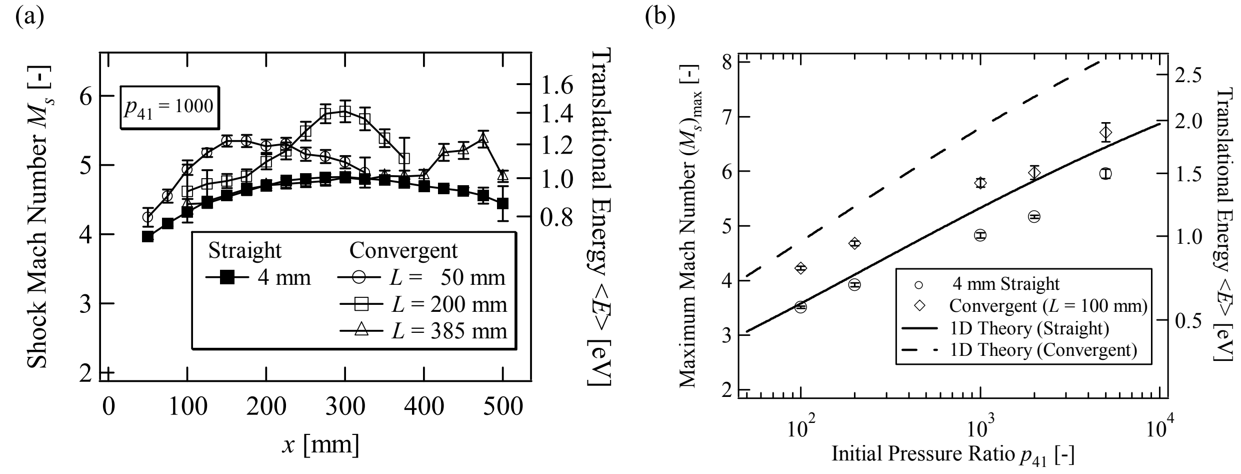

FIGURE 5. Shock Mach numbers in the shock tubes converging linearly in the section of $L \leq x \leq L + 100$ mm. (a) Shock Mach numbers M_s in the convergent tubes with three different geometries (L = 50, 200, and 385 mm) at p_{41} = 1000 as a function of the distance x from the main valve. (b) Maximum Mach numbers $(M_s)_{max}$ in the tube with L = 100 mm as a function of the initial pressure ratio p_{41}.

convergent shock tubes of which diameters linearly decrease from 4 mm to 2 mm. The results indicate that a convergent tube could generate higher-energy molecular beam than straight one by optimizing the position of the convergent section and the tube length.

High frequency operation is indispensable for practical use of the shock-heated molecular beam source. We will improve the exhaust valve to reduce the evacuation time between each shot, and install the shock tube into an existent molecular beam apparatus for performance evaluation.

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