Mechanism of laser and rf plasma in vibrational nonequilibrium CO–N₂ gas mixture

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This paper investigates the mechanism of plasma created by focused CO laser and rf electric field. The plasma is created in a CO/N₂ environment, at a total pressure of 600 torr. Ionization of the gases occurs by an associative ionization mechanism, in collisions of two highly vibrationally excited molecules. These highly vibrationally excited states are populated by resonance absorption of the CO radiation followed by anharmonic vibration-vibration (V-V) pumping. Moreover N₂ also becomes vibrationally excited due to collisions with vibrationally excited CO. The coupled rf reduced electric field E/N is sufficiently low to prevent electron impact ionization that may create plasma individually, so when a subbreakdown rf field is applied to the plasma, collisions between the free electrons heated by the field and the diatomic species create additional vibrational excitation both in the region occupied by the CO laser beam and outside of the laser beam region. The numerical results show plasma created in both regions (in and out of the CO laser beam region) with the associative ionization mechanism. This suggests a method for creating a stable nonequilibrium plasma. The calculation result is verified by comparison the synthetic spectrum to a measured one.

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I. INTRODUCTION

It has always been a great interest for researchers to create and sustain a high-pressure, low-temperature, and large-volume plasma. At high pressures, a positive feedback between gas heating and electron impact ionization rate results in a rapid ionization instability development.¹,² Basically, small electron density perturbations, producing excess Joule heat and thereby increasing the reduced electric field, E/N, result in a more rapid electron generation by the applied field which eventually leads to runaway ionization and glow-to-arc transition in high-pressure gas discharges. The use of external ionization, such as an electron beam sustained discharges, allows decoupling the rate of ionization from the applied electric field, which extends the plasma stability limits to higher pressures and electron densities. However, at high e-beam currents this type of discharge also becomes unstable since the rate of ionization by the beam is inversely proportional to the number density so that gas heating by the beam eventually produces ionization instability. This suggests that a discharge sustained by an external ionization source with a negative feedback between gas heating and ionization rate, and using a subbreakdown electric field to draw the discharge current may be stable at very high pressures.³

An ionization process that satisfies the condition mentioned above is the associative ionization in collisions of two highly vibrationally excited molecules,³–⁵

\[ AB(v) + AB(w) \rightarrow (AB)^{*}_{v+w} + e^{-}, \]

In Eq. (1), AB represents a diatomic molecule, and v and w are vibrational quantum numbers. Basically, ionization is produced in collisions of two highly vibrationally excited molecules when the sum of their vibrational energies exceeds the ionization energy. Ionization by this mechanism has been previously observed in CO–Ar and CO–N₂ gas mixtures optically pumped by resonance absorption of CO laser radiation at pressures of P = 0.1–1.0 atm and temperatures of T = 300–700 K.³⁵–⁸ The optical pumping process used in this work consists of two distinct stages. The first stage is the direct excitation of a target species (CO) to low-energy vibrational levels (ν < 10) by resonance absorption of a broadband, continuous wave (cw), liquid nitrogen cooled, and modest intensity (~10–100 W/cm²) CO laser radiation, in a single photon stepwise process

\[ CO(v-1) + h\nu \rightarrow CO(v). \] (2)

The second stage is the anharmonic collisional intramode vibration-vibration (V-V) up-pumping process,

\[ CO(v) + CO(w) \rightarrow CO(v-1) + CO(w+1), \] (3)

where v and w are vibrational quantum numbers, w > v. The up-pumping is driven by the anharmonicity of the intramolecular potential, which results in process (3) being exothermic in the forward direction. Detailed balance then requires that the forward rate of process (3) exceeds the reverse rate so that the molecule with the larger initial quantum number, w, is preferentially excited by the forward V-V energy transfer process. Adding other diatomic species to the optically pumped CO, such as N₂, also initiates intermode vibration-vibration (V-V') energy transfer, such as

 Eb + Ew > Eion. (1)
Since the energy relaxation rate from the vibrational modes of CO and N\textsubscript{2} remains sufficiently slow, these processes produce strong vibrational excitation of all three diatomic species, CO, N\textsubscript{2} in a low translational temperature, and high-pressure gas mixture sustained by continuous wave CO laser irradiation, at complete steady state.\textsuperscript{5} Under these conditions, ionization of CO occurs by the associative ionization mechanism of Eq. (1).\textsuperscript{6–12} Ultimately, the degree of vibrational excitation in these optically pumped nonequilibrium plasmas, a gas temperature rise results in a more rapid relaxation of the upper vibrational level and their depopulation.\textsuperscript{13} In other words, ionization by mechanism of Eq. (1) can be limited and even terminated by the gas heating. While this method can create high-pressure, low-temperature optically pumped plasmas, the plasma is confined to the relatively small volume defined by the incident CO laser beam. Outside the CO laser beam the electrons rapidly recombine, while diffusion of the vibrationally excited species is limited due to the high pressure. Ploenjes \textit{et al.}\textsuperscript{1} showed that applying an external rf field to the optically pumped plasma at $P\sim1$ atm significantly increased both the plasma volume and the vibrational temperature on the laser beam center line, without creating plasma instabilities. A qualitative scenario of the mechanism of vibrational energy coupling from the rf field to the optically pumped plasma has also been suggested. The main objective of the present paper is to develop a kinetic model of the rf energy coupling process to the optically pumped plasma.

**II. KINETIC MODEL**

In Fig. 1, the schematic geometry of a model shows a CO laser (about 10 W) is used to irradiate a gas mixture which is nitrogen containing 1% of carbon monoxide at a pressure of $P=600$ torr. Two 3 cm diameter brass plate electrodes are mounted approximately 13 mm apart so that the laser beam creates a roughly cylindrical excited region between the electrodes. A rf power is coupled through the probe electrodes. A two-dimensional (2D) time-dependent model is developed to simulate the previously described optically pumping processes as well as the rf power coupling. The model incorporates the equations for electron and positive ion concentrations, as well as the Poisson equation for the electric field,

$$\frac{\partial n_k(x,y,t)}{\partial t} + \nabla \Gamma_k = Q_k,$$

$$\Gamma_k = -D_k \nabla n_k(x,y,t) - \frac{q_k}{\epsilon} \mu_k \nabla \phi(x,y,t)n_k(x,y,t),$$

$$n_k(x,0,t) = n_k(x,d,t) = 0,$$

$$\frac{\partial n_k(0,y,t)}{\partial x} = \frac{\partial n_k(\pm W,y,t)}{\partial x} = 0,$$

$$n_k(x,y,0) = 0,$$

$$\nabla^2 \phi(x,y,t) = \frac{1}{\epsilon_0} \sum_k q_k n_k(x,y,t),$$

$$\phi(x,0,t) = 0, \quad \phi(x,d,t) = U_0 \cos(2\pi \nu t),$$

$$\frac{\partial \phi(0,y,t)}{\partial x} = \frac{\partial \phi(\pm W,y,t)}{\partial x} = 0,$$

$$\phi(x,y,0) = U_0 y/d,$$

$$Q_e = S - \beta n_n,$$

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In Eqs. (5)–(7), the index $k$ stands for electrons and positive ions, $e$ is the electron charge, $n_k$ and $q_k=\pm e$ are the species concentrations and charges, $D_k$, $\mu_k$, and $\Gamma_k$ are the species diffusion coefficients, mobilities, and fluxes (conduction current density vectors), respectively, $\phi$ and $\vec{E}=-\nabla \phi$ are the electric potential and electric field, $j=\Sigma q_j \Gamma_k$ is the electric current, $d$ is the separation between the electrodes, $W$ is the electrode half width, $U_0$ and $\nu$ are the applied voltage amplitude and frequency, $S$ is the rate of electron production by the associative ionization mechanism of Eq. (1), and $\beta$ is the electron-ion recombination coefficient.

Equations (5)–(7) are accompanied by equations for vibrational energies of two diatomic species, CO and N\textsubscript{2}, as well as the translational-rotational energy equation

$$\frac{\partial e_{\text{vib}}(x,y,t)}{\partial t} - \nabla \left[ D_j \nabla e_{\text{vib}}(x,y,t) \right] = R_{\text{elas}} + R_{\text{field}} - R_{\text{rel}},$$

$$e_{\text{vib}}(x,0,t) = e_{\text{vib}}(x,d,t) = e_{\text{vib}}(\pm W,y,t) = e_{\text{vib}}(T_0),$$

$$\frac{\partial e_{\text{vib}}(0,y,t)}{\partial x} = 0,$$

$$e_{\text{vib}}(x,y,0) = e_{\text{vib}}(T_0).$$
\[
\frac{5}{2}kN \frac{\partial T(x,y,t)}{\partial t} - \nabla [\lambda \nabla T(x,y,t)] = kN(y_{CO}R_{CO}^{\text{rel}} + y_{N_2}R_{N_2}^{\text{rel}}),
\]

\[
T(x,0,t) = T(x,d,t) = T(\pm W,y,t) = T_v, \quad \frac{\partial T(0,y,t)}{\partial x} = 0,
\]

\[
T(x,y,0) = T_v.
\]

In Eqs. (8) and (9), \(e_{\text{vib},j}\) is the average vibrational energy per diatomic molecule (CO or \(N_2\)), the index \(j\) stands for CO or \(N_2\), \(k\) is the Boltzmann constant, \(N\) is the number density, \(y_{CO}\) and \(y_{N_2}\) are mole fractions of CO and \(N_2\) in the mixture, and \(R_{\text{las}}^v\), \(R_{\text{field}}^v\), and \(R_{\text{ele}}^v\) are the rates of vibrational excitation by the laser and electron impact and vibrational energy relaxation, respectively.

It is well known that the vibrational distribution functions (VDFs) of diatomic species at the conditions of extreme vibrational disequilibrium sustained by the combined laser and electric field excitation can be strongly non-Boltzmann. Determination of the vibrational level populations of CO and \(N_2\) in spatially nonuniform, strongly vibrationally excited plasmas, \(f_{v,j}\), would therefore require solution of master equation, i.e., approximately 100 coupled 2D partial differential equations of the general form for every vibrational level of each diatomic species in the mixture, \(j\),

\[
\frac{\partial f_{v,j}(x,y,t)}{\partial t} + \nabla [D_{v,j} \nabla f_{v,j}] = \left( \text{laser excitation} \right)_{v,j} + \left( \text{electron impact} \right)_{v,j} + (V - T)v_j + (V - T)v_j + (V - E)v_j,
\]

\[
f_v = \frac{1}{f_0} \left\{ \begin{array}{ll}
\exp \left( - \frac{E_v T_v}{T} + \frac{\Delta E_v(v-1)}{T} \right), & v \leq v_0 = \frac{1}{2} + \frac{T}{2x\tau}, \\
\exp \left( - \frac{\Delta E_v}{T} \right) \frac{v_a - v}{v_a - v_0}^{1/2} & v_0 + 1, \\
- \frac{T}{12\Delta E_v} \sum_M y_M P_{10}^M \exp \left( \frac{\delta_{10}^M(v)}{v+1} \right), & v_0 < v < v_a.
\end{array} \right.
\]

In Eq. (10), \(f_{v,j}\) are the relative populations of vibrational levels (VDF) and \(v\) is the vibrational quantum number. The explicit expressions for the terms in the right hand side of Eq. (10) can be found in Ref. 8. For the conditions where the spatial nonuniformity of the vibrationally pumped gas has cylindrical symmetry (e.g., across a cylindrical pump laser beam), both the time-resolved and the steady-state solutions of Eq. (10), depending on a single spatial coordinate in the radial direction, have been obtained in Refs. 8 and 14. However, for the conditions of the present paper, i.e., in the presence of the external rf field, the solution of Eq. (10) depends on two spatial coordinates \((x, y)\), which makes direct integration of Eq. (10) rather computationally intense. In addition, the rf field oscillation period \((-10^{-7} \text{ s})\) is much shorter compared to the time for vibrational relaxation \((-10^{-4} - 10^{-3} \text{ s})\), which imposes an additional constraint on the integration time step.

For this reason, in the present work the kinetic model retains only two equations for the vibrational energy modes of CO and \(N_2\), \(e_{\text{vib},j}\) [Eq. (10)], obtained from the master Eq. (10) using the relations

\[
e_{\text{vib},j} = \sum_v e_{v,j} f_{v,j}, \quad e_{v,j} = \omega_{v,j} \nu[1 - x_{v,j}(v + 1)],
\]

where \(\omega_{v,j}\) and \(x_{v,j}\) are the vibrational frequency (in K) and the anharmonicity of the molecules. The VDFs of CO and \(N_2\), \(f_{v,j}\), have been determined using approximate analytical theory of vibration-vibration (V-V) pumped anharmonic oscillators,\(^\text{13}\) as a steady-state solution of master Eq. (10) without spatial diffusion terms,

\[
\tilde{Q}_{10} = Q_{10}^S + \frac{\delta_{10}^V}{12 \Delta V} \left( \frac{\pi}{\Delta V} \right)^{1/2} Q_{10}^L,
\]

\(y_M\) is the V-T relaxation collision partner mole fraction, and \(v_a = E_a/E_1\) is the CO vibrational level which corresponds to a near resonance of a rapid vibrational energy transfer process,

\[
\text{CO}(v_a) + N_2(w = 0) \rightarrow \text{CO}(v_a - 2) + N_2(w = 2),
\]

\(v_a = 38\). Equation (12) describes the steady-state nonequilibrium VDF, \(f_{v,j} \sim 1/(v+1)\), formed by the near-resonance V-V exchange processes CO–CO and truncated by the “one-for-
two” V-V’ exchange process of Eq. (16) and by the V-T relaxation. Note that at the present conditions (low-temperature CO–N2 mixtures) the effect of the V-T processes on the VDF is insignificant, and the process of Eq. (16) is the dominant process truncating the CO VDF at the high vibrational levels.

The analytic VDFs of Eq. (12) are in good agreement with steady-state distribution functions obtained by numerical integration of master equation with the V-T and V-V rates given by Eqs. (13) and (14). In the present calculations, the two VDFs have been determined iteratively from Eq. (11) using the functional dependence Eq. (12) on the first level vibrational temperatures, Tνj, as adjustable parameters. This approach greatly reduces the number of equations solved while taking into account the key effect of strongly non-Boltzmann distribution function on the associative ionization rate in the plasma. This approach assumes that both vibrational distributions are quasi-steady-state and that their perturbation by diffusion is relatively weak.

The rate of CO vibrational excitation by resonance absorption of the CO laser radiation, used in Eq. (8), is evaluated as follows: \[ R_{\text{CO}}^{\text{las}} = \sum_{\nu, j} \frac{A_{\nu} B_{\nu}}{8 \pi c k T} \left( \frac{c}{\nu} \right)^3 I_\nu \delta S_j \exp \left( - \frac{B_{\nu}(J + 1)}{T} \right) \epsilon_{\nu, j} \varepsilon_{\nu, v}. \] (17)

where Aνν is the Einstein coefficient for spontaneous radiation, Bν is the rotational constant, ν is the laser line frequency, Iν is the laser line power density, θ is the absorption line shape factor, J is the rotational quantum number, and Sj is the Hönig–London factor.

The rate of vibrational excitation of CO and N2 molecules by electron impact in the presence of the rf field is estimated as follows:

\[ R_{\text{vib}}^{\text{field}} = \frac{\tilde{J} \cdot \tilde{E}}{N k}. \] (18)

The approximation of Eq. (18) is applicable in the range of the reduced electric field E/N = (0.5–5.0) × 10^{-6} V cm², where more than 95% of the input electric power, \( \tilde{J} \cdot \tilde{E} \), goes to vibrational excitation of CO and N2.

The rate of energy relaxation from the vibrational mode of each diatomic species with the VDF given by Eq. (12) is evaluated as follows:

\[ R_{\text{vib}}^{\text{rel}} = E_j \sum_{\nu} \frac{6 \bar{O}_{10} \Delta E}{T \delta_{YY}} (v + 1)^2 \left( 1 - \frac{2 \nu^3 \Delta E}{T} \right) \left( 1 - \frac{E_j - E_0}{T} \right) \left( f_0 f'_0 - f_0 f'_j \right) N. \] (19)

In Eq. (19), the first term in the right hand side represents the intramode energy relaxation rate due to V-T and V-V processes combined, while the second term is the energy relaxation rate due to the V-V’ energy exchange between vibrational modes of CO and N2.

\[ \text{CO}(v) + \text{N}_2(v') \rightarrow \text{CO}(v-1) + \text{N}_2(v' + 1). \] (20)

Again, Eq. (19) is in good agreement with the steady-state energy relaxation rate obtained from the numerical solution of the master equation.

In the gas discharge plasma, if there exists a small amount of dissociated N- or O-atom, they may change the vibrational relaxation processes via V-T relaxation process with collisions with the dissociated atoms. Nevertheless, at current conditions, the low gas temperature and small amount of dissociated N- or O-atom (because of the sub-breakdown electric field, energy more than 95% goes into vibrational mode) determine that the V-T relaxation process of N2 molecule by N-atom is slow. At the same time, considering the difficulty of calculation of N-atom concentration, the effect of dissociated atoms on VDF of molecules is neglected in this paper.

The electron production rate by the associative ionization mechanism of Eq. (1), used in Eq. (7), is calculated as follows:

\[ S = k_{\text{ion}} N^2 \sum_{E_i, E_f} f_{E_i, CO} f_{E_f, CO}. \] (21)

The electron and ion mobilities and diffusion coefficients in a nitrogen plasma as functions of the reduced electric field E/N, where N = P/kT is the number density, are taken from Refs. 15 and 16. The V-V and V-T rates in CO–N2 mixture are taken from Refs. 8 and 17. The CO pump laser spectrum is taken to be the same as in Ref. 18.

Direct numerical solution of coupled partial differential Eqs. (5), (6), (8), and (9) remains rather problematic due to the fact that the characteristic time scales for reaching the quasi-steady-state distributions of electric field, \( \tau_{\text{rf}} \sim d / w_{\text{dr}} \), vibrational populations and electron density, \( \tau_{\text{vib}} \sim \tau_{\text{ion}} \sim E_i / R_{\text{el}}^2 \), and gas temperature, \( \tau_{\text{HT}} \sim d^2 / D \), are widely different.

\[ \tau_{\text{rf}} \sim 10^{-6} \text{ s} < \tau_{\text{vib}} \sim \tau_{\text{ion}} \sim 10^{-3} \sim 10^{-3} \text{ s} \ll \tau_{\text{HT}} \sim 1 \text{ s}. \] (22)

In the above estimates, d ~ 1 cm, \( w_{\text{dr}} = \mu_p E / 10^6 \) cm is the electron drift velocity, and D ~ 1 cm²/s is the diffusion coefficient. The values of \( \tau_{\text{vib}} \) for CO and N2 are estimated from Eqs. (12) and (19) for \( T = 300 \text{ K}, T_{\text{e}}(\text{CO}) \sim 3000 \text{ K}, N \sim 10^{18} \text{ cm}^{-3}, \gamma_{\text{CO}} \sim 0.01, \) and \( \gamma_{\text{N}_2} \sim 1 \). For this reason, first the rms electric field distribution has been evaluated by integrating Eqs. (5) and (6) separately, over the time period \( \Delta t \sim \tau_{\text{rf}} \), and then assumed to remain unchanged while integrating Eqs. (8) and (9) over \( \Delta t \sim \tau_{\text{vib}} \sim \tau_{\text{ion}} \). These two integration stages were repeated until the steady-state spatial distribution of rms electric field, rms charged species concentrations, vibrational level populations, and gas temperature was achieved over \( \Delta t \sim \tau_{\text{HT}} \).

In the calculations discussed in Sec. III, the 3 cm diameter circular electrodes (see Fig. 1) are modeled as square electrodes of the same surface area. The electrode separation and full width are \( d = 1 \text{ cm} \) and \( 2w = 2.66 \text{ cm} \), respectively. The CO laser beam power distribution is assumed to be Gaussian with the \( 1/e^2 \) radius of 1 mm. The calculations have been done for a condition of a CO–N2 mixture with 1%
of CO at $P=600$ torr. The rf voltage amplitude and frequency are $U_0=2750$ V and $\nu=10$ MHz, respectively. The system of Eqs. (5), (6), (8), and (9) is solved using standard differential method.19

III. RESULTS AND DISCUSSION

The results of modeling calculations using the kinetic model described in Sec. II are presented as follows. Figure 2 shows contour plots of the first level vibrational temperatures of CO and N$_2$, as well as the translational temperature and the electron number density in optically pumped CO/N$_2$ plasmas. The vibrational temperature is defined by Eq. (23),

$$T_v = \frac{\theta_1}{\ln[f_0/f_1]}.$$  \hspace{1cm} (23)

Here, $\theta_1=3353$ K is the energy of the first vibrational level of N$_2$, and $f_0$ and $f_1$ are the relative populations of vibrational levels $v=0$ and $v=1$, respectively. The left half of each of these plots refers to the parameter distribution in the plasma sustained by a CO laser alone, while the right half shows distribution of the same parameters in the plasma sustained by a combination of the laser and the rf field. For the laser/rf sustained plasma, the shown parameters have been averaged over the rf field oscillation period. From Fig. 2, it can be seen that applying the rf field to the optically pumped plasma produces a stronger vibrational disequilibrium, with vibrational temperatures of both CO and N$_2$ considerably increasing (2800–4150 K for CO, 1900–3020 K for N$_2$ on the laser beam centerline). With the rf field on, the N$_2$ vibrational temperature exceeds $T_v=1500$ K, and the CO vibrational temperature exceeds $T_v=2000$ K at most locations. Also, N$_2$ vibrational temperature is much higher than the translational temperature at all locations. In these situations, rf energy is coupled to the optically pumped plasma by heating of free electrons formed in the optically pumped plasma by the associative ionization process of Eq. (1), with subsequent vibrational excitation by heated electron impact and gas heating in vibrational relaxation processes. Note that with the rf field turned on the size of the vibrationally non-equilibrium region also substantially increases.

From Fig. 3, which shows a contour plot of the reduced electric field, $E/N$, it can be seen that the highest rms value of $E/N$ in the plasma does not exceed $E/N\approx 1.7$.
$10^{-16}$ V cm$^2$, which is far too low for the rf field induced electron impact ionization at present condition. For this reason, the rf field does not directly contribute to ionization in the plasma. The mechanism of electron number density increase in the plasma away from the laser beam is associative ionization of CO, vibrationally excited by the rf field, by the mechanism of Eq. (1). The negative feedback between the vibrational excitation and the rate of associative ionization is provided by the vibrational energy relaxation from the vibrational mode of CO due to $V$-$V$ exchange CO–CO and $V$-$V'$ exchange CO–N$_2$, given by Eq. (19). This mechanism limits the amount of rf power coupled to the plasma and increases the translational temperature, which self-stabilizes the plasma and precludes the ionization-heating instability development.

Synthetic CO first overtone infrared emission spectra under the condition of rf field turned on and off are plotted in Fig. 4 and compared to a measured infrared emission spectroscopy from Ref. 1. The left plot is experimentally measured and the right one is synthesized from calculation. Both of them shows that coupling of the rf field to the optically pumped plasma can significantly increase the infrared emission signal intensity (by a factor of 7), and the agreement between measured spectra and synthetic spectra is good. Spatially averaged CO VDFs, which correspond to synthetic spectra in Fig. 4, are shown in Fig. 5. It can be seen that the high vibrational level populations considerably increase when the rf field is turned on. It is clear that much more CO molecules are pumped to higher vibrational levels when rf field is applied.

Calculations of the energy balance in the plasma shows that the absorbed laser power in the region between the electrodes is 0.32 W, while the rf power coupled to the plasma is greater by more than an order of magnitude, 4.3 W (see Fig. 6). More rapid vibrational energy relaxation in V-V exchange processes in the laser/rf sustained plasma produces additional heat removed from the plasma by conduction. Diffusion of vibrationally excited molecules provides another channel of energy removal from the plasma. In the present calculations, convective cooling of the gas mixture in the slow-flow absorption cell is neglected. From Fig. 2, one can see that the calculated translational temperature on the laser beam axis increases from $T=320$ K in the laser-sustained plasmas to $T=650$ K in the laser/rf sustained plasma.

**IV. CONCLUSION**

The modeling calculation has been carried out in present work for a high-pressure (600 torr) CO/N$_2$ gas mixture. It is clear that a nonequilibrium plasma can be created and sustained by a combination of a continuous wave CO laser and a subbreakdown radio frequency electric field. The numerical results also show that applying the rf field to an optically pumped CO/N$_2$ plasma produces stronger vibrational disequilibrium both in the region occupied by the CO laser beam and outside of this region. The calculation result is verified by comparison the synthetic spectrum to the previous measured spectrum.
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