Research article

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Temperature-dependent dark-field scattering of single plasmonic nanocavity

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Abstract: Plasmonic materials have long been exploited for enhanced spectroscopy, integrated nanophotonic circuits, sensing, light harvesting, etc. Damping is the key factor that limits their performance and restricts the development of the field. Optical characterization of single nanoparticle at low temperature is ideal for investigating the damping of plasmons but is usually technically impractical due to the sample vibration from the cryostat and the surface adsorption during the cooling process. In this work, we use a vibration-free cryostat to investigate the temperature-dependent dark-field scattering spectroscopy of a single Au nanowire on top of a Au film. This allows us to extract the contribution of electron-phonon scattering to the damping of plasmons without performing statistics over different target nanoparticles. The results show that the full width at half-maximum of the plasmon resonance increases by an amount of 5.8%, over the temperature range of 5–150 K. Electromagnetic calculations reveal that

the temperature-insensitive dissipation channels into photons or surface plasmon polaritons on the Au film contribute up to 64% of the total dissipations at the plasmon resonance. This explains why the reduction of plasmon linewidth seems small at the single-particle level. This study provides a more explicit measurement on the damping process of the single plasmonic nanostructure, which serves as basic knowledge in the applications of nanoplasmonic materials.

Keywords: dark-field spectroscopy; electron-phonon damping; gold nanowire; low temperature; plasmonic nanocavity.

1 Introduction

The local electromagnetic field around a metallic nanostructure can be enhanced by orders of magnitude due to the excitation of localized surface plasmon resonance (LSPR) [1–3], a phenomenon that has been widely used in enhanced spectroscopy [4–6], sensing [7–9], and strong light-matter interaction [10–12], etc. Propagating along the metal-dielectric interface, surface plasmon polaritons (SPPs) are capable of carrying information via their light degrees of freedom, which has been demonstrated for integrated nanophotonic circuits [13, 14]. However, plasmonic materials inevitably suffer from remarkable energy dissipation arising from both radiative and non-radiative damping [15, 16]. The former converts plasmons into free-space photons; the latter annihilates plasmons and creates electron-hole excitations via electron-phonon scattering [17], electron-surface scattering [18] and electron-electron scattering [19]. These damping channels jointly determine the lifetime of surface plasmons, ranging from a few femtoseconds to tens of femtoseconds. While, among these processes, the electron-phonon scattering is the only one that remarkably depends on the temperature [20]. Thus, investigating electron-phonon scattering damping variation with temperature is crucial to either understanding the basic dissipation physics or promoting further developments of the plasmonic applications.

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Plasmon damping significantly broadens the LSPR linewidth of single nanoparticles, providing a robust way to characterize the temperature-dependent damping. A common method to study the LSPR linewidth of nanoparticles is to measure their full width at half-maximum (FWHM) of the LSPR-modulated photoluminescence (PL) from the materials [21–23] or the dark-field (DF) scattering spectra [15, 24–27]. Although the PL method usually has a higher signal to noise ratio, due to its easily eliminated excitation background. It has several limitations. First, it only works for metals with a broad and featureless luminescence spectrum, like Au. The spectral range that this method can apply is restricted by the metal itself. Even for Au, it becomes impractical in the near-infrared region (>800 nm). Second, the heating effect from the laser becomes non-negligible, especially when the metals are less luminescent [28]. Third, the LSPR-modulated PL spectrum will show a blue shift compared to the scattering spectrum, particularly in the short-wavelength regime [29, 30]. It will deform the lineshape of the PL spectrum and perturb the measurement of the FWHM. Technically, FWHM measured from the low-temperature DF scattering can break the above-mentioned limitations, yet, with the lower signal-to-noise ratio. This is because of the adsorption of air, water vapor and dust-motes in the cryostat, onto the sample surface and the chamber window with the decreasing temperature. The vibrations from a typical liquid helium-cooled cryostat usually result in the defocus and drift of the sample under the microscope, increasing the difficulty of tracking the same nanoparticle throughout the experiment. As a result, previous studies on temperature-dependent plasmon damping had to perform statics over different nanoparticles [20, 26], which in principle brings in inhomogeneous broadening. *In situ* low-temperature DF measurements on one single plasmonic nanostructure are key to remove the uncertainty.

In this paper, we use a vibration-free cryostat to investigate the temperature-dependent DF scattering spectroscopy on one single plasmonic nanostructure. This drift-free measurement allows us to track a single nanostructure to extract the contribution of electron-phonon scattering without statistics from numerous different nanoparticles at different temperatures. To increase the signal-to-noise ratio of the low-temperature DF spectroscopy, we chose a plasmonic structure with a strong polarization response and added polarizers into the setup. The structure consists of a Au-nanowire (Au-NW) over mirror (NWOM), which supports cavity plasmons in the gap region, similar to other nanoparticles over mirror systems [31–39] that receive intense interest. The use of cavity

plasmons benefits from their directional emission toward the substrate normal that improves the collection efficiency of the scattered light.[40] The choice of NW, instead of nanocubes or nanodisks, significantly increases the total scattering signal since its length is much longer. Also, the 1D NW makes the NWOM highly anisotropic in the substrate plane, allowing the introduction of polarization microscopy. Analogous to the conventional Fabry-Pérot (FP) cavity, the NWOM nanocavity can be viewed as a truncated metal-insulator-metal waveguide that supports numerous orders of cavity plasmons [41], whose energy can leak from the nanocavity and transmit into propagating SPPs [42] in the direction perpendicular to the NW (analogous to the photons leaking from FP cavity).

2 Materials and methods

2.1 Materials and sample preparation

The cetyltrimethylammonium bromide (CTAB) wrapped Au-NWs used in the experiment were synthesized via a seed growth approach [43], and the ultrasmooth Au film was fabricated by the template-stripping method [44]. A 5-nm thick aluminum oxide layer was deposited onto the ultrasmooth Au film by atomic layer deposition at 200 °C. The thickness of this dielectric layer defined the gap distance g between the NW and the film. To construct the NWOM, Au-NWs (dispersed in ethanol) were dropped onto the Au film. And the sample was washed with acetone and deionized water to remove the surfactant layer capped on the Au-NWs. Then, the sample was dried by nitrogen flow. Finally, another aluminum oxide layer (thickness $t = 5$ nm) was deposited on the top of the NWOM to isolate it from the ambient environment in the cryostat. This step reduced the effect of surface adsorption and thermal expansion on the plasmon peak positions during the cooling process.

2.2 Optical characterization

The temperature-dependent DF scattering spectrum of Au-NWOMs was measured with a Horiba spectrometer (iHR550). The sample was mounted on a liquid helium-free cryostat (Motana, Cryostation s50 – CO) that operated with a very small vibration. An obliquely incident (45°) white light from a halogen lamp (Olympus) was focused onto the sample. A polarizer was placed behind the lamp to realize the polarization-adjustable excitation. The scattered light from NWOM was collected by a $50\times$ DF objective (Olympus, NA = 0.5). An analyzer was placed after the object to analyze the polarization of the collection. The scattered signal passing through a beam splitter (R:T = 5:5, Thorlabs) was either collected by the spectrometer or captured by a CCD (Q-IMAGING, ROLERA EM-C2). Finally, the scanning electronic microscopy (SEM) was used to characterize the morphologies of the NWOM and confirm the dimensions of the NWOM. The experimental scattering spectra were post-processed by subtracting the background spectrum collected from a nearby region.

2.3 Numerical calculation

Basic geometry. The electromagnetic simulations were performed by the FEM method (COMSOL Multiphysics 5.2a). In the simulations, the dielectric function of the Au followed the data from Johnson&Christy [45], and the refractive index of aluminum oxide and SiO₂ was 1.5. Considering the system symmetry, a 2D geometry was built. The Au-NW was simulated as a 62 nm-diameter pentagonal rounded by a 4 nm curvature, situated on a 200 nm Au film with 5 nm aluminum oxide spacer in between. Depositing a 5 nm conformal aluminum oxide layer on the top of whole geometry, the NWOM model was finally created. A 200 nm thick PML layer covered the whole geometry.

Far-field transformation: scattering and coupling to SPPs. Excited by a 45 degrees oblique plane wave, the energy of far-field radiation (scattering) and near-field SPPs excitation was calculated using an open-source near-to-far field transformation package.[42] Like every near-field to far-field transformation, a closed transformation surface surrounded the inhomogeneities (scatters) of the system. Thus, the equivalent principle [46], treating the far-field radiation from the scatters equivalently as the radiation from the electric/magnetic surface current sources on the transformation surface, can be applied to investigate the far-field diagram. However, the Green function – the relationship between near-field current sources and far-field radiation is hard to obtain in our NWOM system with stratified bottom layers. In this open-source package [42], the reciprocity theory was applied [42, 46]. Instead of solving the Green function of stratified layers analytically, test wave from every polar angle (2D coordinate) was inversely applied to calculate the responsive currents on the transformation surface, which would correspond to the Green function of the system due to the reciprocity. Therefore, the far-field scattering could be finally obtained. For the same reciprocity principle, the energy of the guided mode could also be calculated [42].

Absorption of the NWOM. The absorption of the system at ~700 nm (M mode) has two origins: the LSPR and SPPs. The absorption of SPPs is equal to the energy of the guided mode calculated from the far-field transformation. It is a straightforward result due to the exponential decay of the SPPs, and there are no SPPs in the infinitely far field. The absorption due to the LSPR (localized absorption) consists of two components – Au-NW and Au film. The absorption of the Au-NW Q_{nw}^{M}

was easily obtained by surface integrating of $\text{Im}(\epsilon)|E|^2$, while the absorption of Au film should be carefully identified because the total Ohmic loss consisted of both contributions from SPPs Q_{SPPs} and LSPR $Q_{\text{film}}^{\text{M}}$. Following our previous work [35], the contribution of LSPR could be fitted out from the absorptions of the gradually increasing semicircle integral domain in the Au film (details see Supplementary S1). When the integral domain is larger than the localized LSPR hotspot, the relationship between increasing absorption corresponding to the radius is linear (see Supplementary S1). By applying a linear fitting to the specific range of absorption data, the intercept of the plot should be the localized absorption owing to the LSPR $Q_{\text{film}}^{\text{M}}$. The localized absorption of the NWOM was defined as $Q_{\text{M}} = Q_{\text{nw}}^{\text{M}} + Q_{\text{film}}^{\text{M}}$.

3 Results and discussion

In the experiment, we used a home-made oblique-incidence DF microscopy combined with a vibration-free cryostat to investigate the temperature-dependent DF scattering spectroscopy of a single Au-NWOM system (Figure 1A). Oblique illumination can improve the signal-to-noise ratio of the DF scattering signal, and the vibration-free cryostat can avoid the drift of the target NWOM during the cooling process (detail setup in Method 2.2). Figure 1B shows the cross-section of the NWOM configuration with a diameter of the NW $d \sim 62 \pm 2$ nm, the thickness of the ultrasmooth gold film $h \sim 200$ nm, the spacer Al₂O₃ ~5 nm, and the deposited Al₂O₃ layers protection $t \sim 5$ nm. The simulation result (modeling details in Method 2.3) is shown in Figure 1C: the far-field scattering spectrum, collected by a 0.5 NA cone, contains two characteristic plasmonic modes. The T mode at ~528 nm arises from the transverse dipole mode of the NW, with the electric field distributions shown in Figure 1D. The M mode ~700 nm originates from

Figure 1: (A) Optical setup for the DF scattering spectra measurements (B) Schematic of a single NWOM system: a crystalline Au-NW ($d = 62 \pm 2$ nm) with a pentagonal cross-section, situated on an ultrasmooth gold film separated by a 5-nm-thick Al₂O₃ spacer ($g = 5$ nm). Another 5-nm-thick Al₂O₃ layer is coated on the top of the NWOM ($t = 5$ nm). (C) Calculated scattering spectrum of NWOM shown in (B). Two resonant peaks correspond to the transverse dipole mode (T) and the lowest-order cavity plasmon mode (M). (D),(E) The electric field distributions of the T mode and M mode.

the magnetic plasmon resonance [41], which could also be recognized as the lowest order nanocavity mode [47]. The electric field is greatly enhanced in the gap region under the M mode (Figure 1E). The simulation well accords to the experiment DF scattering shown in Figure 2D and F.

To elucidate the polarization dependence of the DF scattering in the NWOM system, we changed the excitation and detection polarization, respectively, at room temperature. As shown in Figure 2A and B, the DF optical images of NWOM under different polarized illumination show a dramatic color contrast – lively red under horizontal excitation, and totally dark under perpendicular excitation. It means that the magnetic mode was efficiently excited through horizontal excitation (Figure 2A, E field parallel to the NW short-axis), while the perpendicular excitation (Figure 2B, E field parallel to the NW long-axis) has no LSPR in the probing wavelength range. Figure 2C is the SEM image of the NW demonstrated in Figure 2A and B, whose partially magnified view is shown in the inset. To understand the polarization dependence of the system quantitatively, we collected a series of scattering spectra under the different polarized excitations. The definitions of the excitation angle θ and detection angle α are shown in the inset of Figure 2D. They are the angles between the polarization and the long-axis of the NW. We first fixed $\alpha = 90^\circ$, meaning that the direction of detection polarization is perpendicular to the long axis of the NW. Then the polarizer in the excitation light path was rotated to change the excitation angle to investigate the excitation polarization dependence. Figure 2D proves that the LSPR modes (T and M) are sufficiently excited when $\theta = 90^\circ$ corresponding to the bright DF image in Figure 2A. However, $\theta = 0^\circ$ shows no

obvious signals, corresponding to the dark DF image in Figure 2B. The intensities of M mode (orange spheres in Figure 2E) are collected against the excitation angle θ , revealing that the DF scattering intensity was strongest as $\theta = 90^\circ$ and weakest as $\theta = 0^\circ$ (180°). It indicates a strong polarization dependence of the nanocavity mode (M mode). When the polarization is perpendicular to the long axis of the NW ($\theta = 90^\circ$), surface plasmons will propagate back and forth in the truncated MIM configuration to form a standing wave (nanocavity mode) [41, 8, 47]. The lowest-order nanocavity mode (M mode) supported by the short axis shows a distinct scattering intensity in the visible regime, while M mode for the long axis is supposed to be at the mid-infrared. That is why we found the $\theta = 0^\circ$ DF intensity orders of magnitude weaker than that of $\theta = 90^\circ$. Besides, we found the intensities in Figure 2E fitting well with the relation: $I(\theta) = \sin^2 \theta$, which reveals good dipolar behavior in excitation.

Likewise, the detection polarization-dependence of the DF scattering is shown in Figure 2F and Figure 2G. We fixed the excitation polarization angle $\theta = 90^\circ$ and rotated the analyzer in the detection path. Similar dipolar relation was observed in Figure 2G with the maximum detection signal at $\alpha = 90^\circ$ and the minimum at $\alpha = 0^\circ$ (180°). It means that the nanocavity plasmon radiation has a dipole-shaped emission diagram, and the radiation is at the maximum when the detection angle is perpendicular to the NW. The above experimental results indicate that the NWOM system shows good dipolar behavior both in light excitation and radiation. To get the strongest scattered signal, both the excitation polarization and the detection polarization should be perpendicular to the long axis of the NW.

Figure 2: Polarization-dependent DF optical images of the NWOM systems under (A) horizontal excitation (B) perpendicular excitation. (C) SEM image of an NWOM with a detailed SEM image of the end of the Au-NW in the inset. Inset scale bar: 100 nm. (D) and (F) represent the excitation- and detection-polarization dependent scattering, respectively. θ (α): the angle between excitation (detection) polarization and the long axis of Au-NW, respectively. The relative peak intensities as a function of excitation and detection polarization are shown in the (E) and (G), respectively, exhibiting a good dipole behavior in light excitation and scattering.

We performed the temperature-dependent experiment and recorded a series of DF spectra at various temperatures (5–150 K). In order to get the maximum detection efficiency, both the excitation polarization and the detection polarization were rotated to be perpendicular to the long axis of the NW. Because the geometry of the Au-NWOM system is well protected by a layer of conformal aluminum oxide, the LSPR wavelength shift from 5 K (703 nm) to 150 K (706 nm) is only 3 nm demonstrated in Figure 3A. This small shift means that this measured Au-NWOM system is stable. The linewidth under each temperature was extracted from the DF spectra by Lorentzian fitting, the specific relationship between the FWHM broadening and the temperature is presented in Figure 3B. Those orange spheres indicate the values of FWHM of the M mode under each temperature. The FWHM of these spectra was broadened with the increasing temperature from 5 to 150 K, which accords with the damping process of electron-phonon scattering. In low temperatures, fewer phonons will be excited, which would decrease the electron-phonon scattering probability and extend the mean free path of electrons as well as the lifetime.

To quantitatively describe the plasmon damping process [48], the total plasmon damping rate Γ_t , proportional to the total linewidth of the M mode, can be separated into three terms: the radiative decay into photon Γ_{rad} , the coupling to SPPs Γ_{SPPs} , and the local Ohmic heating $\Gamma_{\text{abs}}(T)$. The first two terms are assumed to be temperature independent because both of them are determined by the impedance matching that is governed solely by electromagnetism (Maxwell equations). The effects of temperature physically enter these electromagnetic processes through the permittivity of the materials. If neglecting the thermal expansion on both the structural parameters and the permittivity, the temperature will only affect the electron-phonon coupling that changes the imaginary part of

the permittivity. According to the Kramers-Kronig relation, the real part of the permittivity will also be altered, resulting in the change of coupling to photons and SPPs. One should keep in mind that the temperature-dependent electron-phonon coupling is the only physical origin. The variation of the SPPs coupling efficiency against the temperature is the consequence of the reduction of Ohmic absorption, not the origin of it. Therefore, the temperature-dependent total plasmon damping rate can be written as: $\Gamma_t(T) = \Gamma_{\text{rad}} + \Gamma_{\text{SPPs}} + \Gamma_{\text{abs}}(T)$. Although the SPPs will eventually attenuate into Ohmic heating, this subsequent SPPs absorption will not affect the FWHM of the M mode because the SPPs will propagate irreversibly away from the NWOM. The propagation distance of SPP on the Au surface is typically around 10 μm , much larger than the length scale of the near field of the M mode. This is why the temperature-dependent SPPs propagation cannot be repeatedly taken into consideration, but the coupling efficiency between the M mode and the SPPs does. Thus, the heat dissipation in this NWOM system merely comes from the localized absorption of the LSPR $\Gamma_{\text{abs}}(T)$. Meanwhile, the localized absorption damping channel contains contributions from electron-phonon scattering, electron-electron scattering, and electron-surface scattering: $\Gamma_{\text{abs}}(T) = \Gamma_{\text{e-ph}}(T) + \Gamma_{\text{e-e}} + \Gamma_{\text{e-surf}}$. In this equation, the electron-phonon induced damping is the only term related to the temperature variation. The ratio η of the temperature-dependent heat loss to the total heat dissipation in this system follows $\eta = \Gamma_{\text{e-ph}}(T)/\Gamma_{\text{abs}}(T)$.

In order to explain the temperature-dependent dephasing from the DF, a solid-state Debye model, describing the relationship between electron-phonon relaxation and temperature, is applied. Since the occupied states of electrons and phonons start to change with the interband transitions, the solid-state Debye model is only applicable in the absence of the interband transitions. The $\Gamma_{\text{e-ph}}(T)$ is given by [20]:

$$\Gamma_{\text{e-ph}}(T) = \frac{\hbar}{\tau_0} \left[\frac{2}{5} + 4 \left(\frac{T}{\Theta} \right)^5 \int_0^{\Theta/T} \frac{z^4}{e^z - 1} dz \right] \quad (1)$$

In this formula, the Θ and τ_0 are two constants for the given metal material. τ_0 is a constant related to the near-infrared absorption of the metal, which equals to 30 fs for gold in our configuration [26]. Θ represents the Debye temperature (170 K) of the metal, according to the previous study [49]. The 2/5 term arises from the spontaneous phonons radiation, relating to the electron delay process in the metal nanostructure. When the temperature is below 50 K, this part is the minimum of the electron-phonon interaction, which is 8.8 meV for gold nanostructure [20]. The term

Figure 3: (A) DF scattering spectra of a NWOM with temperature varying from 5 K to 150 K. (B) The measured FWHM (orange dots) and calculated dephasing time (solid black line) of the M mode at different temperatures. The dephasing time was calculated by the Debye model.

on the rightmost represents the dependence of the phonon modes on the temperature. This solid-state Debye model can simulate the electron-phonon scattering process when the temperature of the sample is below the Debye temperature of 170 K.

The solid black line in Figure 3B is the relaxation time derived from the low-temperature Debye model Eq. (1), which illustrates the electron-phonon scattering influence on the FWHM (decay rate or the lifetime, equivalently). From Figure 3B, we can see that the low-temperature Debye model can well account for our experimental results extracted from the FWHM, proving that the temperature-dependent homogeneous broadening of the LSPR linewidth (or plasmon damping) is originated from the electron-phonon scattering. Consistent with what we have qualitatively demonstrated in the theoretical model, the FWHM has very little variation at the temperature lower than 50 K, which indicates that the temperature-dependent decay of the plasmon is a constant in this temperature range. According to the fitting of the DF scattering spectra (see Supplementary S2), the FWHM of the M mode increases by approximately 5.8%, from 155.5 to 164.5 meV as the temperature increases from 5 to 150 K. The total dephasing time of the plasmon can be calculated by $\tau = 2\hbar/\Gamma$, $\hbar$ is the reduced Planck constant. The results showed that the dephasing time decreases from 8.5 fs to 8 fs when the temperature was increased from 5 to 150 K. We also explored the relationship at higher temperatures range (150 ~ 246 K, see Supplementary S2), which also shows the FWHM gradually homogeneous broadening as the temperature increases.

The plasmon damping process is accompanied by energy dissipation. In this NWOM system, dissipation due to electron-phonon interaction mentioned above is a component of the LSPR absorption energy loss channel. Apart from this channel, the energy dissipation during plasmon decay into photons and SPPs launching should also be considered. Therefore, in this NWOM system, energy is dissipated in three forms: far-field scattering, SPPs launching, and localized absorption (see Method 2.3). As shown in Figure 3A, the peak position is slightly affected by the temperature (the shift is ~ 3 nm). Thus we believe that the geometry of NWOM is stable as the temperature changes. Therefore, the percentage of each energy loss component in the NWOM is constant over our selected temperature range (5 – 150 K). To study the proportion of each channel participating in the plasmon damping, we calculate the relative intensity of each energy loss component, as is shown in Figure 4A (calculation details see Method 2.3). The black line represents the total power loss of the system, containing the components of localized

absorption from LSPR (red line), far-field scattering (orange line), as well as the excitation of SPPs on the interface (blue line). Overall, three peaks are observed, with two modes already demonstrated in previous mode identifications (Figure 1C–E). A second-order nanocavity mode shoulder around 600 nm, which was not observed in our experiment due to the low object NA (Figure 2D and F), was predicted in the calculation. This mode was excited following the retardation excitation mechanism realized by the oblique incident excitation. Overall, we find the maxima of the energy dissipation on resonances, indicating that those dissipation channels are all highly dependent on the LSPR. Three dissipation channels are comparable at the resonance wavelength, indicating that the M mode has both intense radiation and absorption. In the range of 570 – 680 nm, the second-order nanocavity mode dominates the energy dissipation. As for the second-order nanocavity mode, due to the mode symmetry, the even-order nanocavity modes tend to be darker and less coupled with the plane waves due to the poor field overlap, which accounts for the minor shoulder in the far-field scattering. However, the weak far-field scattering, in return, gives a relatively high SPPs excitation efficiency because it is the only leaking channel for the surface plasmons confined in plasmonic nanocavity. The local absorption of LSPR (red line) plays a major role in all decay channels below 570 nm due to the interband transition. The imaginary part of the dielectric constant is higher in this range, which improves the absorption efficiency. Figure 4B shows the energy loss component proportion of each channel. On resonance, the proportions of the plasmon energy dissipation channel into localized absorption is around 36%, and temperature-independent channels – the power of far-field radiation and SPPs are around 33% and 31%, respectively. We also examined the temperature-dependent Drude model [50] to

Figure 4: (A) Simulated intensity of each energy component during the plasmon dissipation. The black line represents the total energy losses, including the energy loss from localized absorption (red line), far-field scattering (orange line), and the SPPs excitation (blue line). (B) The percentage of the three energy components. The gray dashed lines in two figures indicate the resonant wavelength of the LSPR (700 nm).

calculate the energy portions at low temperatures (see Supplementary S3), which shows a slight redistribution of the energy due to the temperature-dependent absorption. This large proportion of temperature-independent channels ($\sim 64\%$) accounts for the small dephasing time variation against the temperature change (Figure 3B). Combined with the experimentally fitted FWHM at 150 K, the dissipation values of the three channels are 59.2 meV (localized absorption), 54.3 meV (far-field scattering), and 51 meV (SPPs), respectively.

To quantitatively investigate the dissipation mechanism in the range of 5–150 K, we calculated the decay rates of different channels. $\Gamma_{e-ph}(T)$ can be derived from the total damping rate subtracted by the temperature-independent damping rate Γ_0 : $\Gamma_{e-ph}(T) = \Gamma_t(T) - \Gamma_0$, where the Γ_0 is the sum of the Γ_{SPPs} , Γ_{rad} , Γ_{e-e} and Γ_{e-surf} . After the derivation procedures (shown in Supplementary S4), we conclude that the temperature-independent damping is 146.7 meV at 5 K. And the electron-phonon coupling damping could be calculated, which is 17.8 meV at 150 K. For a specifically-designed plasmonic structure, energy dissipation caused by far-field scattering and SPPs coupling can be eliminated, but the energy dissipation due to the absorption (Ohmic loss) is inevitable. As a result, the percentage (η) of electron-phonon induced heat dissipation to the total heat dissipation is a more important parameter since the percentage of $(1-\eta)$ represents the ultimate lower limit of the total loss rate for a given plasmonic structure which can not be eliminated by adjusting the parameters of the structure. Also, it is geometry-independent to allows a fair comparison when discussing the effect of temperature on the Ohmic loss in different geometric nanostructures. For example, we compare this percentage (η) in this work with that of the others work. According to the calculation in Supplementary S4, the ratio η of temperature-dependent heat dissipation, namely $\Gamma_{e-ph}(T)$, to the total heat dissipation $\Gamma_{abs}(T)$ is 30.1% at 150 K. A slightly larger value of 34.3 % was obtained in another NWOM sample under the same condition (shown in Supplementary S5), indicating that the results are reliable. The η of NWOM in our experiment is overall lower than that of the single Au nanorods, $\eta \sim 37\%$ at 153 K, reported by Alexander Konrad et al. The reason for this variation mainly arises from the geometric difference between the NWOM nanostructure and the nanorods. Because of the mean-free path of the conduction electrons varies with the size and shape of the nanostructure, the influences of the electron surface scattering are different in this two experiments which result the variation of the η . Besides, Alexander Konrad et al. carried out their measurement by statistics over different nanorods at each temperature, which inevitably causes the

inhomogeneous broadening of the FWHM. We uses the same NW to measure the FWHM broadening at each temperature, which effectively overcomes the large FWHM error caused by the statistical method. Moreover, the value of $\Gamma_{abs}(T)$ in our experiment is obtained by combining the calculated percentage of the absorption with the measured total FWHM of the LSPR. Inevitable calculation errors may affect the accuracy of the results.

In fact, using temperature to modulate the plasmon resonance energy, linewidth, and amplitude for realizing the plasmon activation have been widely studied. Previous work mainly used a combination of tunable refractive index materials and plasmonic structures to tune the plasmons by temperature, such as the investigation on the hybrid of plasmonic-phase changing materials and plasmon structures [51–52], the thermally controlled liquid crystal molecules to realize the active control of plasmons [53], thermo-optical materials such as PMMA to be used in active plasmonic structures [54]. In addition to changing the dielectric surroundings of the plasmonic nanostructure, there are two other ways to control plasmon resonance actively: by controlling the gap distance of the plasmonic structure, [8] and using a self-tunable the plasmon structure [55]. This self-tunability of noble metals can be achieved by controlling their dielectric functions or inherent carrier densities. Our experiment uses the self-tuning capability of Au to adjust the resonance linewidth by changing the temperature without the assistance of thermo-sensitive materials. It not only provides an explicit measurement method for a single plasmonic structure under low temperature but also enriches the investigation of the active plasmonics.

4 Conclusion

In conclusion, we investigated the plasmonic dissipation mechanism by performing an *in situ* temperature-dependent DF experiment on a single NWOM system via a homemade oblique-incidence single-particle DF spectroscopy combined with a vibration-free cryostat. Different from the conventional studies probing different nanoparticles at different temperatures, we carried out the whole low-temperature experiment on one single NWOM, which eliminates various uncertainties and ambiguity induced by the individual differences from various nanoparticles. We demonstrated the DF scattering properties of a single NWOM system at the temperature ranging from 5–150 K and derived the temperature-dependent dephasing time. Due to electron-phonon scattering, the FWHM of the DF scattering spectra was found to be broadened by 5.8%

when the temperature was increased from 5 to 150 K. At the temperature of 150 K, the percentage (η) of the temperature-dependent heat dissipation to total heat dissipation is 30.1%. The numerical calculations verified that the temperature-insensitive dissipation channels into photons or SPPs on the Au film contribute up to 64%. This study promotes the understanding of the damping mechanism in a single plasmonic nanostructure and serves as a guide for future applications.

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