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Spin resonance and high-frequency optical properties of the cuprates

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We argue that recently observed superconductivity-induced blue shift of the plasma frequency $\delta\omega_{pl}$ in $Bi_2Sr_2CaCu_2O_{8+\delta}$ [Molegraaf *et al.*, Science **295**, 2239 (2002)] is related to the change in the integrated dynamical structure factor associated with the development of the spin resonance below T_c. We show that the magnitude of $\delta\omega_{pl}$ is consistent with the small integrated spectral weight of the resonance, and its temperature dependences closely follow that of the spin-resonance peak. We also discuss the differential optical integral for the conductivity.

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The importance of the resonance spin mode for the physics of the cuprates continue to be the subject of intensive debate. In a generic superconductor, the pairing of fermions drastically reduces the damping of collective spin degrees of freedom at energies below 2Δ. For a d-wave superconductor, the residual interaction between spin fluctuations and fermions gives rise to the additional effect—the development of the exciton mode below 2Δ (see, e.g., Ref. 1, and references therein). This mode exists for bosonic momenta near (π,π) and is commonly called the "spin resonance." It has been observed in three different families of high-T_c superconductors.[2–5] This mode is not a "glue" to superconductivity as it emerges only in the superconducting state (more precisely, below the pseudogap temperature), but it affects electronic properties of the cuprates in the superconducting state.

Much of recent works on the effect of the spin resonance on electrons was concentrated on whether the interaction with the resonance is capable to explain experimentally detected *low-energy* features in the fermionic spectral function, tunneling density of states, and optical conductivity.[1,6] An example of this behavior is the peak-dip-hump structure of the spectral function.[7]

The subject of the present communication is the analysis of the possible role of the spin resonance in the observed changes between normal and superconducting state in the optical data at high frequencies, $\omega \sim 0.5-1$ eV.[8] Recently, Molegraaf *et al.* reported the results of their ellipsometry measurements on optimally doped and underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ with $T_c=88$ and 66 K, respectively.[9] They observed that in the normal state the in-plane plasma frequency increases with decreasing T, roughly as T^2, however below T_c it increases faster, and the actual value of $\omega_{pl}(T=0)$ is larger than the extrapolation from the normal state. The effect is small: at optimal doping $\delta\omega_{pl} \sim 10$ cm^{-1} is 10^{-3} of the plasma frequency $\omega_{pl} \approx 1$ eV, but detectable by the ellipsometry technique. They and others[10,11] also found that superconductivity affects the temperature dependence of the optical integral $I(\Omega)=\hbar^2\int_0^\Omega \sigma(\omega)d\omega$ at $\Omega \sim 1$ eV.

In this paper, we focus on the changes of ω_{pl} and $I(\Omega)$ between normal and superconducting states, leaving aside the mechanism of the temperature dependence in the normal state. We argue that the superconductivity-induced shifts of the plasma frequency and the optical integral can be explained by the interaction with the spin-resonance mode. We argue that $\delta\omega_{pl}$ and $\delta I(\Omega)$ scale with the integrated magnetic spectral weight, and the small values of $\delta\omega_{pl}$ and $\delta I(\Omega)$ are consistent with the fact that only 1–2% of the magnetic spectral weight is transferred into resonance. We emphasize that this effect is absent in phonon superconductors where the spectral function of the pairing boson does not change below T_c.

Conventional wisdom holds that at $\omega \sim 1$ eV, which well exceed the magnetic bandwidth, interaction with spin fluctuations is not the dominant mechanism for the fermionic self-energy $\Sigma(\omega)$. We argue, however, that $\delta\omega_{pl}$ scales with $\delta\Sigma(\omega)=\Sigma_{sc}(\omega)-\Sigma_n(\omega)$, and the latter comes from frequencies comparable to the superconducting gap and can be captured within the low-energy, spin-fluctuation theory. We will see, however, that at high ω, a fermion with energy 1 eV interacts with the whole band of magnetic fluctuations. As a result, $\delta\Sigma(\omega)$ scales with the integrated magnetic spectral weight transfer into the resonance.

Our reasoning is the following. At plasma frequency, the real part of the dielectric function $\epsilon(\omega)$ changes sign. The dielectric function obeys $\epsilon(\omega)=\epsilon(\infty)+4\pi i\sigma(\omega)/\omega$, where $\sigma(\omega)$ is the optical conductivity. By Kubo formula, $\sigma(\omega)=[(\omega_{pl}^0)^2/(4\pi)]\text{Re}[\Pi(\omega)/(-i\omega)]$ where, $\Pi(\omega)$ is the fully renormalized current-current polarization operator and $(\omega_{pl}^0)^2=4\pi ne^2/m$ is the bare plasma frequency. The plasma frequency is then the solution of

$$\omega_{pl} = \frac{\omega_{pl}^0}{\sqrt{\epsilon(\infty)}}\sqrt{\text{Re}\Pi(\omega_{pl})}. \tag{1}$$

To zero-order approximation, $\Pi(\omega_{pl})=1$, i.e., $\omega_{pl}=\omega_{pl}^0/\sqrt{\epsilon(\infty)}$. However, at any finite frequency $1-\text{Re}\Pi(\omega)$ is still finite, and hence ω_{pl} is sensitive to the change of the polarization operator upon entering the superconducting state. This change of ω_{pl} is small as superconductivity mostly affects the form of $\Pi(\omega)$ at frequencies comparable to the superconducting gap $\Delta \sim 0.04\omega_{pl}$.

At high frequencies, $\omega \sim 1$ eV, normal and anomalous fermionic self-energies $\Sigma(\mathbf{k},\omega)\approx\Sigma(\omega)$ and $\Phi(\mathbf{k},\omega)$ are both small compared to ω, and to the leading order in the self-

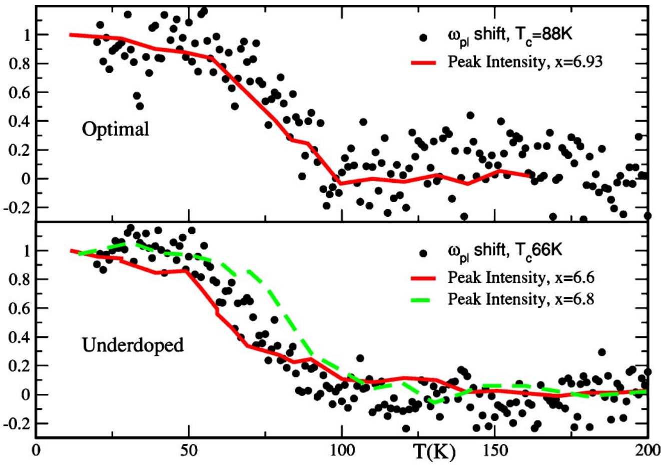

FIG. 1. The temperature dependence of the superconductivity-induced change of the plasma frequency in Bi2212 (Ref. 9) vs the change in the resonance peak intensity for YBCO (Ref. 4). We normalized both quantities to 1 at $T=0$. For ω_{pl}, the normal state value below T_c was obtained by extrapolating the T^2 dependence from the normal state (Ref. 9). Upper panel: optimally doped YBCO; lower panel: two underdoped samples.

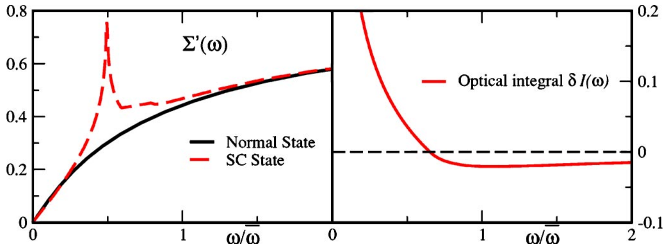

FIG. 2. The results of Eliashberg calculations for coupling $\lambda=1$. Left panel: self-energy. Right panel: the optical integral $\delta I(\omega)$ (including the condensate piece). For $\lambda=2$, $\delta I(\omega)$ changes sign at a larger $\omega\sim 2\overline{\omega}$.

energy, the current-current correlator both in the normal and superconducting states is given by

$$\Pi(\Omega) = \int_0^\Omega \frac{d\omega}{\Omega + \Sigma(\omega) + \Sigma(\Omega - \omega)}. \qquad (2)$$

Vertex corrections to this formula are not dangerous as the scattering of an electron off spin excitation changes the direction of electron's velocity by almost 90°.[6] For such processes transport time does not differ much from a single-particle lifetime.

Substituting Eq. (2) into (1) we find that the superconductivity-induced change in the plasma frequency at $T=0$ is related to the difference between superconducting and normal fermionic self-energies $\delta\Sigma(\omega)=\Sigma^{sc}(\omega)-\Sigma^{n}(\omega)$ as

$$\frac{\delta\omega_{pl}}{\omega_{pl}} = -\frac{1}{2\Pi'_n(\omega_{pl})} \times \mathrm{Re}\int_0^{\omega_{pl}} d\omega \frac{\delta\Sigma(\omega) + \delta\Sigma(\omega_{pl}-\omega)}{[\omega_{pl} + \omega Z_\omega + (\omega_{pl}-\omega)Z_{\omega_{pl}-\omega}]^2}, \qquad (3)$$

where $\delta\omega_{pl}=\omega_{pl}^{sc}-\omega_{pl}^{n}$, $\Pi'_n(\omega)$ is the real part of the polarization operator in the normal state, and $Z_\omega=1+\Sigma_n(\omega)/\omega$ is the inverse quasiparticle residue in the normal state. By all accounts, at $\omega\sim\omega_{pl}$, $\Sigma(\omega)\ll\omega$, i.e., $Z_\omega\approx 1$. Hence, once the integral in (3) is dominated by frequencies where either ω or $\omega_{pl}-\omega$ are near ω_{pl} (as we later verify), the precise form of the fermionic $Z(\omega)$ does not matter, and $\delta\omega_{pl}/\omega_{pl} \approx \delta\Sigma(\omega_{pl})/(2\omega_{pl})$.

The computation of $\delta\omega_{pl}$ therefore reduces to the computation of the self-energy difference between normal and superconducting states. We present the result for $\delta\Sigma(\omega)$ now and discuss its derivation and approximations later. We found that, within the spin-fluctuation scenario, there are two distinct frequency regimes depending, roughly, on whether or not ω exceeds the magnetic bandwidth. At small frequencies, the Eliashberg approximation is valid, and $\delta\Sigma(\omega)$ is positive (see Fig. 2). In this regime, internal fermions and bosons in the self-energy diagram have comparable energies, i.e., a fermion is interacting only with spin fluctuations very near **Q**. If this behavior was valid up to $\omega\sim\omega_{pl}$, $\delta\omega_{pl}$ would be negative, in disagreement with the data. However, we found that at $\omega>(1-2)\overline{\omega}$, where $\overline{\omega}$ (defined below) is comparable to magnetic bandwidth, Eliashberg approximation becomes invalid, and an "anti-Eliashberg" approximation has to be used. In this approximation, *all* spin fluctuations contribute to the self-energy, and we obtained that the change of the fermionic self-energy is proportional to the integrated change of the dynamical spin structure factor,

$$\delta\Sigma(\omega) \approx -\frac{3g^2}{Z_\omega \omega}\int \frac{d\Omega d^2q}{8\pi^3}\sum_i \delta S_i(\mathbf{q},\Omega)F(\mathbf{q}), \qquad (4)$$

where g is the spin-fermion coupling constant estimated to be $g\sim 0.7$ eV,[1,6] and $\delta S_i(\mathbf{q},\Omega)=S_i^{sc}(\mathbf{q},\Omega)-S_i^{n}(\mathbf{q},\Omega)$ is the change of the dynamical structure factor $S(\mathbf{q},\Omega)=\chi''(\mathbf{q},\Omega)[1+\coth(\omega/2T)]$ in even ($i=1$) and odd ($i=2$) channels. The factor $F(\mathbf{q})$ decreases away from **Q** but can be safely approximated by $F(\mathbf{q})=1$ in the narrow momentum range where the resonance is experimentally detectable. The experimentally measured integrated magnetic spectral weight near (π,π) is larger in the superconducting state,[3,4] hence at high frequencies $\Sigma'(\omega)$ is *smaller* in a superconductor. Using this $\delta\Sigma(\omega)$ we find that $\delta\omega_{pl}$ is positive, in agreement with Ref. 9.

The momentum and frequency integral in the r.h.s. of (4) yields $BS(S+1)/3=B/4$, where B is the percentage of the spectral weight redistributed below T_c. Only the odd channel contributes to $\delta S(\mathbf{q},\Omega)$ and 1–2% of the spectral weight from this channel is redistributed,[3] i.e., $B\sim 0.005-0.01$. Substituting $\delta\Sigma(\omega)\approx -3g^2B/(4\omega Z_\omega)$ into the expression for $\delta\omega_{pl}$ and approximating Z_ω by 1 we obtain $\delta\omega_{pl}/\omega_{pl}\sim(1-2)\times 10^{-3}$, in near perfect agreement with the experimental result 1.3×10^{-3}. We emphasize that the agreement is entirely due to the fact that the integrated weight of the resonance is very small; if it was not, the blue shift of the plasma frequency would be much larger. To verify the prefactor, we went beyond estimates and evaluated the full integral in (3) using the normal state expression for Z_ω:[6] $Z_\omega\approx 1$

$+(i\overline{\omega}/\omega)^{1/2}$, where $\overline{\omega}\sim 0.35g$. We obtained almost the same result for $\delta\omega_{pl}$ as above, with the extra prefactor $0.97\approx 1$.

In Fig. 1 we plot the temperature dependence of $\delta\omega_{pl}(T)$ below T_c together with the temperature dependence of the resonance peak intensity.[4] The data for the fully integrated intensity are available for fewer temperatures and show roughly the same T dependence.[4] We see that $\delta\omega_{pl}$ and the resonance peak follow each other, as it should be according to Eq. (4).

We next turn to the conductivity. Molegraaf *et al.*[9] and Santander-Syro *et al.*[10] reported that in optimally doped Bi2212, the optical spectral weight, with the condensate contribution included, integrated up to a frequency of $10\,000\ \mathrm{cm}^{-1}$ (above which one-band description fails), increases below T_c. Molegraaf *et al.* interpreted their result as the evidence that the one-band description of superconductivity is not sufficient, and interband transitions also contribute to the pairing. Boris *et al.*,[11] on the other hand, argued, based on their data, that the normal state optical integral becomes larger than the optical integral in the superconducting state above few hundred meV. A similar conclusion follows from recent measurements by Homes *et al.*[12] The discrepancy between the experimental results is at least partly related to the difficulty of extrapolating the normal state conductivity to $T=0$. Santander-Syro *et al.*[10] argued their conclusions depend on the extrapolation procedure.[10]

Our results are consistent with Refs. 11 and 12. We found in our calculations that at optimal doping, the full optical integral $\delta I(\Omega)=I_{sc}(\Omega)-I_n(\Omega)$ is positive at small Ω, but changes sign at $\Omega\sim 300-500$ meV, and is negative at $\Omega \geqslant 1$ eV. In other words, the actual value of the optical integral $I(\Omega\sim 1\ \mathrm{eV})$ in the superconducting state at $T\ll T_c$ is smaller than $I(\Omega)$, extrapolated from the normal state.

Our reasoning is based on the fact that in one-band model, the f-sum rule must be satisfied, i.e., $I(\Omega=\infty)=0$. Hence, $\delta I(\Omega)=-\hbar^2\int_\Omega^\infty \delta\sigma(\omega)d\omega$. Suppose that $\Omega\sim\overline{\omega}$, i.e., it is at the crossover between Eliashberg and anti-Eliashberg regimes. The integral over $\omega>\Omega$ in the optical integral is over the range where Eq. (4) is valid. Since at high frequencies, $\mathrm{Im}[1/Z(\omega)]\approx -\mathrm{Im}\ \Sigma(\omega)/\omega$ is negative, the high-frequency conductivity $\sigma(\omega)\sim 1/\omega^2\tau(\omega)\propto\Sigma''(\omega)$ is larger in the superconducting state than in the normal state, *extrapolated to T* $=0$. Hence, $\delta\sigma(\omega)$ is positive, the integral of it is also positive, and hence $\delta I(\Omega\sim\overline{\omega})$ is negative. Obviously, $I(\Omega)$ will remain negative for all Ω within the range of anti-Eliashberg approximation.

This reasoning implies that the optical integral should change sign already within Eliashberg approximation. To verify this, we computed the optical integral $\delta I(\Omega)$ at $\Omega \leqslant \overline{\omega}$ within Eliashberg theory.[13,14] We present the results in Fig. 2. We indeed obtained that $\delta I(\Omega)$ changes sign at some frequency $\Omega\leqslant\overline{\omega}$, and is negative at higher frequencies. The frequency where $\delta I(\Omega)$ changes sign increases with underdoping, in agreement with Ref. 10 (see caption of Fig. 2), but theoretically it still remains $O(\overline{\omega})$ even in strongly underdoped materials.[14] Note that this does not contradict the idea that that superconductivity is driven by the decrease of the kinetic energy,[15] as within the Eliashberg approach, the decrease of the kinetic energy also comes from frequencies $\sim\overline{\omega}$.[16]

Our results therefore indicate that at $T=0$, the full optical integral changes sign already at frequencies for which Eliashberg approximation is valid, and then gradually decreases with increasing Ω. We remind in this regard that at weak coupling, the optical integral $\delta I(\Omega)$ changes the sign at $\Omega\leqslant 2\Delta$ and is negative at larger frequencies. Our results indicate that the behavior of $\delta I(\Omega)$ does not change drastically between weak and strong couplings—the frequency where $\delta I(\Omega)$ changes sign increases with coupling strength, but still remains smaller than the fermionic bandwidth. This behavior is similar to that in a dirty BCS superconductor.[17] There, $\delta I(\Omega)$ changes sign below 2Δ in the clean limit, and at $\Omega\tau \approx 0.66$ in the dirty limit. At larger frequencies, $\delta I(\Omega)$ is negative and gradually decreases as Ω increases.

The accuracy of our numerical Eliashberg calculations is not sufficient to compare the contributions to $\delta I(\Omega)$ from Eliashberg and anti-Eliashberg regimes. As a rough estimate, we computed anti-Eliashbereg contribution to $\delta I(\Omega)$ by substituting Eqs. (2) and (4) into the Kubo formula. We obtained $\delta I(\Omega)\approx(2-4)\times 10^{-3}(\mathrm{eV})^2$ for $\Omega\sim 1$ eV. Assuming that the (negative) Eliashberg contribution is of the same order, we obtain $|\delta I|\sim 10^{-3}(\mathrm{eV})^2$ for $\Omega\sim 1$ eV, in agreement with Refs. 9 and 10.

We now describe the calculation of the self-energy, Eq. (4), in some detail. We assume that the fermionic self-energy predominantly comes from the fermion-fermion interaction in the spin channel and can be viewed as being mediated by spin collective modes with momenta near (π,π). The imaginary part of the fermionic self-energy is given by

$$\Sigma''(\mathbf{k},\omega)=\frac{3}{8\pi^3}\int \tilde{g}^2\chi''(\mathbf{q},\Omega)G''(\mathbf{k}+\mathbf{q},\omega+\Omega)\times\left(\tanh\frac{\omega-\Omega}{2T}+\coth\frac{\Omega}{2T}\right)d\Omega d^2q, \qquad (5)$$

where $\tilde{g}$ is the fully renormalized vertex, $\chi(\mathbf{q},\Omega)$ is the full dynamical propagator of the collective mode, and $G(\mathbf{k}+\mathbf{q},\omega+\Omega)$ is the full fermionic Green's function.

In the Eliashberg approximation, vertex corrections can be neglected (i.e., $\tilde{g}=g$), and $G(\mathbf{k}+\mathbf{q},\omega+\Omega)$ can be approximated by free-fermion propagator. The reasoning is that low-energy spin fluctuations are overdamped in the normal state and are slow modes compared to electrons. Hence, an effective Migdal theorem is valid.[6] By the same reason, the momentum integration in Eq. (5) can be factorized—the integration transverse to the Fermi surface involves only fast fermions, while the integration along the Fermi surface is over slow bosonic momenta. Within this computational procedure, one finds that Σ'' jumps at $\Delta+\omega_{res}$ that is the key element of the peak-dip-hump behavior.[1,6]

This approximation is, however, valid only as long as external fermionic frequency ω is smaller than a typical frequency at which the momentum integral of $\delta\chi''(\Omega)$ converges.

At strong coupling, this scale is the effective bosonic bandwidth $\overline{\omega}$ defined such that in the normal state, $\Sigma(\omega)$ becomes less than ω at $\omega>\overline{\omega}$, i.e., spin-fluctuation scattering becomes ineffective. When spin-fermion coupling g is less

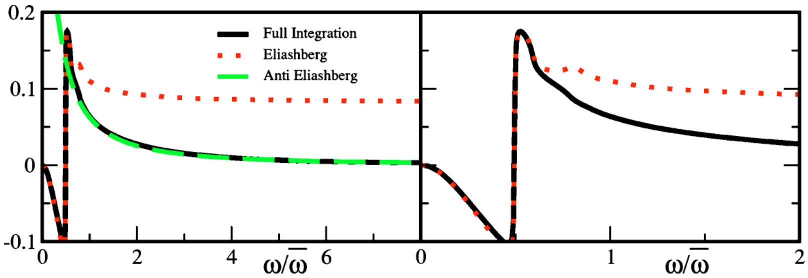

FIG. 3. Left panel: the difference in the electronic self-energy between normal and superconducting states calculated explicitly and using Eliashberg and anti-Eliashberg approximations. The frequency is measured in units of $\overline{\omega} \sim 0.35g \sim 250$ meV. Clearly, the anti-Eliashberg approximation is much better at large frequencies. Right panel: the low-frequency region, where the Eliashberg approximation is valid.

than the fermionic bandwidth W, $\overline{\omega} \sim 0.35\overline{g}$, in the opposite limit $\overline{\omega} \sim W^2/g \sim J$.[6] At larger frequencies, $\omega > \overline{\omega}$, $G''(\mathbf{k}+\mathbf{q}, \omega+\Omega) \approx \text{Im}(\omega Z_\omega)^{-1}$ at $k \approx k_F$, and it can be taken out of the integral. To the same accuracy, the T-dependent factor in (5) is approximated by $1+\coth(\Omega/2T)$. We then obtain

$$\text{Im}[\delta\Sigma(\omega)] = -3g^2 \,\text{Im}\left[\frac{1}{Z_\omega \omega}\right] \int \frac{d\Omega d^2q}{8\pi^3} \sum_i \delta S_i(\mathbf{q},\Omega) F(\mathbf{q}), \tag{6}$$

where $F(\mathbf{q})$ subject to $F(\mathbf{Q})=1$ decreases at $|\mathbf{Q}-\mathbf{q}| = O(|\mathbf{Q}|)$ and reflects the fact that the spin-fermion model is only valid for bosonic momenta near (π,π). As the Kramers-Kronig transform of (6) is infrared convergent, the full self-energy is given by (4).

We see therefore that at high frequencies $\omega \gg \overline{\omega}$, the correct computational procedure for $\delta\Sigma(\omega)$ is opposite to the Eliashberg approximation—instead of factorizing the momentum integral, one can neglect the momentum dependence in the Green's function and perform the full 2D momentum integration over the bosonic momenta. We explicitly verified that in this anti-Eliashberg approximation, vertex corrections are again small, this time in $\overline{\omega}/\omega$, such that $\tilde{g}=g$ in Eq. (5).

Obviously, there should be a crossover between Eliashberg and anti-Eliashberg approximations as frequency increases. To understand where it is located, we evaluated $\delta\Sigma''(\omega)$ explicitly, using the normal and superconducting forms of the dynamical spin susceptibility obtained earlier,[6] and compared the full result with the two approximate forms. The results are presented in Fig. 3. We see that for $\omega \leqslant \overline{\omega}$, Eliashberg approximation is much closer to the full result. However, for $\omega > (1-2)\overline{\omega} \approx 250-500$ meV the Eliashberg approximation is well off, while the anti-Eliashberg approximation is rather close to the full expression. This justifies our use of Eq. (5) for optical properties above 500 meV. Indeed, using the one-band model at these energies we assume that there is a frequency range above $\overline{\omega}$, where anti-Eliashberg approximation is valid, but interband transitions still can be neglected. The applicability of this approximation has to be verified only by comparing the results of one-band analysis with the data, as we did.

To summarize, in this paper we considered the superconductivity-induced blue shift of the plasma frequency and the change of the optical integral. We argued that both can be explained within the magnetic scenario for the cuprates. We found that $\delta\omega_{pl}$ and the differential optical integral $\delta I(\Omega)$ scale with the change of the integrated magnetic spectral weight $\delta S(\mathbf{q},\Omega)$. The relative magnitudes of $\delta\omega_{pl}$ and $\delta I(\Omega)$ are small, $\sim 10^{-3}$, as the integrated $\delta S(\mathbf{q},\Omega)$ accounts for only a small fraction of the total spectral weight. We found that $\delta\omega_{pl}$ is positive, and that $\delta I(\Omega)$ changes sign below 0.5 eV and is negative at larger frequencies.

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