Coulomb Suppression of NMR Coherence Peak in Fullerene Superconductors

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(Received 15 October 1997)

The suppressed NMR coherence peak in the fullerene superconductors is explained in terms of the dampings induced by the Coulomb interaction between conduction electrons. The Coulomb interaction, modeled in terms of the on-site Hubbard repulsion, is incorporated into the Eliashberg theory of superconductivity with its frequency dependence considered self-consistently at all temperatures. The vertex correction is also included via the method of Nambu. The frequency dependent Coulomb interaction induces the substantial dampings in the superconducting state and, consequently, suppresses the anticipated NMR coherence peak of fullerene superconductors as found experimentally. [S0031-9007(98)06592-2]

PACS numbers: 74.70.Wz, 74.20.Fg, 74.25.Nf

It is generally accepted that the superconducting properties of the fullerene superconductors can be understood in terms of phonon-mediated s-wave pairing [1–4]. The $1/(T_1T)$ for s-wave superconductors, where T_1 is the nuclear spin-lattice relaxation time and T is the temperature, is expected to show a peak as T is lowered below the transition temperature T_c and is constant above T_c. It is referred to as coherence peak or Hebel-Slichter peak. The expected coherence peak is found substantially suppressed for fullerene superconductors [5–7]. The underlying mechanism of the suppression, however, is not clearly understood yet. The present Letter addresses the problem of NMR coherence peak suppression in fullerene superconductors and explains it in terms of the dampings in the superconducting state induced by the frequency dependent screened Coulomb interaction between conduction electrons.

The maximum of the normalized relaxation rate, R_s/R_n, in the limit of zero applied magnetic field was estimated to be 1.1–1.2 for fullerene superconductors [6,7], where $R = T_1^{-1}$ is the relaxation rate, and the subscripts s and n, respectively, refer to the superconducting and normal states. A similar behavior was also observed in muon spin relaxation experiments [8]. An analysis based on the phonon-mediated Eliashberg theory [9] gives $T_c/\omega_{\mathrm{ph}} \approx 0.2$, and the characteristic phonon frequency $\omega_{\mathrm{ph}} \approx 100\ \mathrm{cm}^{-1}$ [7]. This seems inconsistent with the commonly held view of $\omega_{\mathrm{ph}} \sim 1000\ \mathrm{cm}^{-1}$ for fullerene superconductors [1–4], which implies $(R_s/R_n)_{\max} \sim 2-3$ [9]. The increase of $(R_s/R_n)_{\max}$ relative to its normal state value, 0.1–0.2, is therefore suppressed by an order of magnitude from the expected value. Closely related is the observation of the unexpected sensitivity of R_s/R_n to the applied magnetic field. The coherence peak is found completely suppressed at only around 5 T for Rb_2CsC_{60}, which is at least an order of magnitude smaller than the expected value [7]. It may be understood on the ground that the zero field coherence peak is substantially suppressed as mentioned above. A simple weighted average of the suppressed intrinsic rate from the superconducting region and the normal rate from the normal region inside the vortex core will give the experimentally observed field dependence of R_s/R_n [7].

In order to understand the coherence peak suppression at zero magnetic field, let us list the possible factors that are known to affect the NMR coherence peak of R_s/R_n [10,11], and check if any of those can explain the suppression. The NMR coherence peak suppression may be attributed to (a) momentum anisotropy of the superconducting gap including non-s-wave pairing, (b) time reversal symmetry breaking such as magnetic impurities or applied magnetic field, and/or (c) the damping effects in the superconducting state. For fullerene superconductors, the superconductivity is of the phonon-mediated s-wave pairing, and, due to the orientational disorder of C_{60} molecules, the Fermi surface anisotropy is not strong enough to suppress the coherence peak [11]. The time reversal symmetry breaking cannot explain the suppression either because there are no magnetic impurities in the fullerene superconductors, and we are considering the case of the zero applied magnetic field. Because either the gap anisotropy or time reversal symmetry breaking cannot explain the coherence peak suppression in the fullerene superconductors, the near absence of the coherence peak should be due to the *damping effects*.

The dampings of an electron come from the scatterings of the electron with the phonons, impurities, and/or other electrons. The damping from the scatterings of electrons with the phonons, that is, the electron-phonon interactions, is not strong enough: The dimensionless electron-phonon coupling constant λ of the fullerenes is estimated to be 0.5–1 [2–4], far smaller than ~2 needed to suppress the NMR coherence peak [12]. The far infrared reflectivity measurements of DeGiorge *et al.* [13] show that the ratio $2\Delta_0/k_BT_c \approx 3.44-3.45$ for both K_3C_{60} and Rb_3C_{60}. It is very close to the BCS value of 3.52 and implies that λ cannot be as large as 2. The impurity scatterings smear out the Fermi surface anisotropy and cannot suppress the coherence peak as explained previously. There remains, therefore, only one possibility for inducing the dampings required to suppress the NMR coherence peak in the fullerene superconductors: the scatterings between electrons due to the *Coulomb interactions*. This idea is indeed verified in our detailed Eliashberg-Nambu (EN) calculations as will be detailed below.

The NMR relaxation rate for a superconductor with a finite bandwidth of B is given by

$$\frac{1}{T_1 T} \propto \int_0^\infty d\epsilon \, \frac{\partial f_F(\epsilon)}{\partial \epsilon} \times \left\{ \left(\mathrm{Re} \frac{\epsilon \theta(\epsilon)}{\sqrt{\epsilon^2 - \Delta(\epsilon)^2}} \right)^2 + \left(\mathrm{Re} \frac{\Delta(\epsilon)\theta(\epsilon)}{\sqrt{\epsilon^2 - \Delta(\epsilon)^2}} \right)^2 \right\}, \tag{1}$$

where $f_F(\epsilon) = 1/(1 + e^{\beta\epsilon})$ is the Fermi distribution function, $\beta = 1/k_B T$, $\theta(\epsilon) = \tan^{-1}[B/2Z(\epsilon) \times \sqrt{\epsilon^2 - \Delta(\epsilon)^2}\,]$, and $Z(\omega)$ and $\Delta(\omega)$ are, respectively, the renormalization and gap functions. The finite conduction bandwidth with a constant density of states (DOS) is explicitly considered through the factor of θ, which is $\pi/2$ for the usual case of infinite bandwidth superconductors. For fullerene superconductors, the Fermi energy $\epsilon_F = B/2 \approx 0.2$–$0.3$ eV and the average phonon frequency $\omega_{\mathrm{ph}} \approx 0.05$–$0.15$ eV. Consequently, $\omega_{\mathrm{ph}}/\epsilon_F \sim 1$ for fullerenes unlike conventional metals, where $\omega_{\mathrm{ph}}/\epsilon_F \ll 1$. When $\omega_{\mathrm{ph}}/\epsilon_F \sim 1$, the phonon vertex correction becomes important because the Migdal theorem does not hold, and the Coulomb interaction should be considered more carefully because the validity of the Coulomb pseudopotential, $\mu^* = \mu/[1 + \mu \ln(\epsilon_F/\omega_{\mathrm{ph}})]$, is unclear. In the present work concerned with the narrow bandwidth superconductor of fullerenes, therefore, the vertex correction is incorporated into the Eliashberg theory via the method of Nambu [14]. The Coulomb interaction is modeled in terms of the on-site Hubbard repulsion and is included in the theory with its frequency dependence considered self-consistently. As far as we are aware, this is the first self-consistent calculation of $1/(T_1 T)$ with the Eliashberg formalism including the Coulomb interaction and the vertex correction. The frequency dependence of the screened Coulomb interaction, which comes from the polarization diagrams of both the normal and pairing processes of renormalized electrons, is important because the electron-phonon and Coulomb interactions vary on a comparable frequency scale for a narrow bandwidth superconductor of $\omega_{\mathrm{ph}}/\epsilon_F \sim 1$.

The NMR coherence peak below T_c is due to the increased DOS in the superconducting state, that is, due to the smallness of the denominator of Eq. (1) when $\epsilon \approx \Delta(\epsilon)$. In order for the peak to be suppressed, therefore, the vanishingly small denominator should be avoided. The $\sqrt{\epsilon^2 - \Delta(\epsilon)^2}$ of Eq. (1) may not vanish when there is a damping, that is, nonzero $\Delta_2(\epsilon)$ for $\epsilon \approx \Delta_1(\epsilon)$, where Δ_1 and Δ_2 are, respectively, the real and imaginary parts of the gap function. The dampings in the superconducting state responsible for the NMR coherence peak suppression is greatly increased when the frequency dependence of the screened Coulomb interaction is retained for $\omega_{\mathrm{ph}}/\epsilon_F \sim 1$. There have been several papers which emphasize the importance of including both the electron-phonon and electron-electron interactions in understanding the fullerenes [15].

The EN equation can be written in the Matsubara frequency as

$$\begin{aligned} Z_n p_n &= p_n + \frac{1}{\beta} \sum_m [\lambda_{\mathrm{ph}}(n-m) - \lambda_{\mathrm{ch}}(n-m) + \lambda_{\mathrm{sp}}(n-m)] \frac{2\theta_m Z_m p_m}{\sqrt{p_m^2 + \Delta_m^2}} + \frac{1}{\pi\tau} \frac{\theta_n p_n}{\sqrt{p_n^2 + \Delta_n^2}}\,, \\ Z_n \Delta_n &= \frac{1}{\beta} \sum_m [\lambda_{\mathrm{ph}}(n-m) - \lambda_{\mathrm{ch}}(n-m) - \lambda_{\mathrm{sp}}(n-m)] \frac{2\theta_m Z_m \Delta_m}{\sqrt{p_m^2 + \Delta_m^2}} + \frac{1}{\pi\tau} \frac{\theta_n \Delta_n}{\sqrt{p_n^2 + \Delta_n^2}}\,, \end{aligned} \tag{2}$$

where $p_n = \pi T(2n + 1)$ is the Matsubara frequency, $\theta_n = \tan^{-1}(B/2Z_n\sqrt{p_n^2 + \Delta_n^2})$, and $\lambda_{\mathrm{ph}}(n - m) = \int_0^\infty d\Omega \, \alpha^2 F(\Omega) 2\Omega/[\Omega^2 + (p_n - p_m)^2]$ is the pairing kernel due to the electron-phonon interaction. Equation (2) is of the same form as the theory used to study the spin fluctuation effects on superconductivity [16]. The $\lambda_{\mathrm{ch}}(n - m)$ and $\lambda_{\mathrm{sp}}(n - m)$ are, respectively, the interactions in the charge and spin channels due to the Hubbard repulsion, and are determined self-consistently as

$$\begin{aligned} \lambda_{\mathrm{ch}}(k) &= UN_F\{1/2 - (\chi_n + \chi_s) + (\chi_n + \chi_s)^2 \\ &\quad \times \ln[1 + 1/(\chi_n + \chi_s)]\}\,, \\ \lambda_{\mathrm{sp}}(k) &= UN_F\{1/2 + (\chi_n - \chi_s) + (\chi_n - \chi_s)^2 \\ &\quad \times \ln[1 - 1/(\chi_n - \chi_s)]\}\,, \end{aligned} \tag{3}$$

where $\chi_n(k)$ and $\chi_s(k)$ are the dimensionless susceptibilities from, respectively, the normal and pairing processes given by

$$\begin{aligned} \chi_n(k) &= \frac{N_F U}{\epsilon_F} \frac{1}{\beta} \sum_l \theta_l \theta_{k+l} \frac{p_l p_{k+l}}{\sqrt{p_l^2 + \Delta_l^2}\sqrt{p_{k+l}^2 + \Delta_{k+l}^2}}\,, \\ \chi_s(k) &= \frac{N_F U}{\epsilon_F} \frac{1}{\beta} \sum_l \theta_l \theta_{k+l} \frac{\Delta_l \Delta_{k+l}}{\sqrt{p_l^2 + \Delta_l^2}\sqrt{p_{k+l}^2 + \Delta_{k+l}^2}}\,. \end{aligned} \tag{4}$$

The Z_m on the right-hand side of Eq. (2) represents the vertex correction of Nambu, which we take as its form in the normal state for simplicity [14]. Including the vertex correction enhances the transition temperature in accord with the previous papers [17]. If we neglect the vertex correction, the Z_m should be put equal to 1. The self-consistent solution of Eq. (2) together with Eqs. (3) and (4) gives $Z(ip_n)$ and $\Delta(ip_n)$ in the imaginary frequency. To obtain $Z(\omega)$ and $\Delta(\omega)$ in the real frequency, we perform the analytic continuations using the iterative method of mixed representations [11,18]. It is more efficient than solving the Eliashberg equation directly in the real frequency. The details of the Eliashberg-Nambu formulation in the imaginary frequency and its analytic continuation will be reported separately.

The EN equation of Eq. (2) is solved self-consistently via iterations with a set of the phonon spectral function $\alpha^2F(\Omega)$, Hubbard repulsion U, and impurity scattering rate τ^{-1}. To model the fullerene superconductors, we take $\alpha^2F(\Omega) = \sum_{\nu=1}^{3} \alpha_\nu^2 F_\nu(\Omega)$, where $F_\nu(\Omega)$ is the truncated Lorentzian centered at ω_ν with the broadening $\Gamma = \omega_\nu/5$, the cutoff frequency $\Gamma_c = 3\Gamma$, and $\int_0^\infty d\Omega\, F_\nu(\Omega) = 1$ [11]. Various theoretical and experimental estimates do not agree well with each other in terms of distribution of coupling strength α_ν^2 among the different modes. These estimates show, however, that the phonon spectra derived from the intramolecular A_g and H_g modes are distributed over 0.03–0.2 eV with the total λ in the range 0.5–1 [2–4]. In view of this, we represent the phonon modes with three groups centered around $\omega_\nu = 0.04, 0.09, 0.18$ eV, and $2N_F\alpha_\nu^2/\omega_\nu = 0.3\lambda_s, 0.2\lambda_s, 0.5\lambda_s$, respectively, for $\nu = 1, 2, 3$. Note that $\sum_{\nu=1}^{3} 2N_F\alpha_\nu^2/\omega_\nu = \lambda_s$. This choice of $\alpha^2F(\Omega)$ gives the logarithmically averaged phonon frequency $\omega_{ln} \approx 0.094$ eV, which is a representative value of the various estimates of ω_{ln}. The λ_s is set to give $T_c \approx 20$–40 K for a given U. We take the Fermi energy $\epsilon_F = B/2 = 0.25$ eV in the present calculations, which gives the ratio $\omega_{ln}/\epsilon_F \approx 0.38$.

We put $\tau^{-1} = 0$ for simplicity because the results are insensitive to the impurity scatterings. Even though the Anderson theorem does not hold exactly because of the finite bandwidth and the Hubbard repulsion in the present theory, the thermodynamic properties are still insensitive to the impurity scatterings. We take $UN_F = 0.31$ and $\lambda_s = 0.71$ in the numerical calculations reported below. We find $T_c = 0.0031$ eV and $2\Delta_0/k_BT_c = 3.1$. It is interesting to note that the present theory gives a rather small $2\Delta_0/k_BT_c$ value which lies at the low end of the various estimates of the gap values. In the present study, the long wavelength contribution of the Coulomb interaction is not considered explicitly. Therefore, U should be taken as a screened value. The previous estimates give $UN_F \approx 0.3$–0.4 [2].

Figure 1 shows the superconducting gap function $\Delta(\omega)$ as a function of ω at $T = 0.001$ eV obtained by solving the Eliashberg-Nambu equation. We took 220 Matsubara frequencies to solve Eq. (2) by iterations and disregarded 20 high frequency data to avoid boundary effects. The analytic continuations were carried out with 2000 frequencies in the range between 0–0.6 eV. Figure 1(a) is for an interacting system of $UN_F = 0.31$ and $\lambda_s = 0.71$, and Fig. 1(b) is for a noninteracting case of $U = 0$ and $\lambda_s = 0.35$, which gives $T_c = 0.0032$ eV and $2\Delta_0/k_BT_c = 3.7$. The solid and dashed lines, respectively, represent the real and imaginary parts of $\Delta(\omega)$.

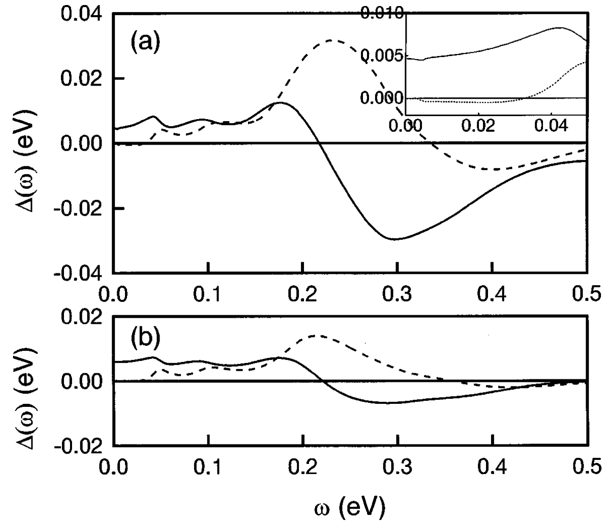

FIG. 1. The superconducting gap function, $\Delta(\omega)$, at $T = 0.001$ eV. The solid and dashed lines, respectively, stand for the real and imaginary parts. We take $UN_F = 0.31$ and $\lambda_s = 0.71$ for (a), and $U = 0$ and $\lambda_s = 0.35$ for (b), so that the two cases have similar T_c. The substantial damping of $|\Delta_2/\Delta_1| \approx 0.05$ around $\epsilon \approx \Delta_1(\epsilon)$ for the interacting case of (a) can be seen more clearly in the inset of (a).

The three peaks in $\Delta(\omega)$ reflect the three peaks in the phonon spectral function $\alpha^2F(\Omega)$. Note the difference in $\Delta_2(\omega)$ between the two cases for $\epsilon \approx \Delta_1(\epsilon)$: For the interacting case (a), the superconducting state has quite a strong damping of $|\Delta_2/\Delta_1| \approx 0.05$ around $\epsilon \approx \Delta_1(\epsilon)$ due to the Coulomb interaction even at the low temperature of $T/T_c \approx 0.3$, while $\Delta_2/\Delta_1 = 0$ around $\epsilon \approx \Delta_1(\epsilon)$ for the noninteracting case of (b) because the thermal fluctuations are quenched at this temperature. As T is increased, $|\Delta_2/\Delta_1|$ is further increased due to the thermal fluctuations. Substantial Δ_2 is what suppresses the NMR coherence peak as shown in Fig. 2.

The $\Delta(\omega)$ and $Z(\omega)$ obtained above are then used to calculate the relaxation rate T_1 using Eq. (1). We show in Fig. 2 the normalized relaxation rate R_s/R_n as a function of the reduced temperature T/T_c. The solid line is for $UN_F = 0.31$ and $\lambda_s = 0.71$ corresponding to Fig. 1(a). As expected, the substantial damping in the superconducting state in the interacting system suppresses the NMR coherence peak so that the maximum of $R_s/R_n \approx 1.15$. By comparison, the corresponding curve for the noninteracting case of Fig. 1(b) ($U = 0$ and $\lambda_s = 0.35$) exhibits a much more pronounced coherence peak as shown by the dotted curve, a hallmark of the weak-coupling s-wave BCS superconductors. It is clear that the strong Coulomb interaction can suppress the NMR coherence peak for phonon-mediated s-wave superconductors with a modest electron-phonon coupling constant. As U is increased, the NMR coherence peak is further suppressed. The dashed curve shows R_s/R_n for $UN_F = 0.4$ and $\lambda_s = 0.8$, which gives $T_c \approx 0.003$ eV. We now show some of the available experimental data

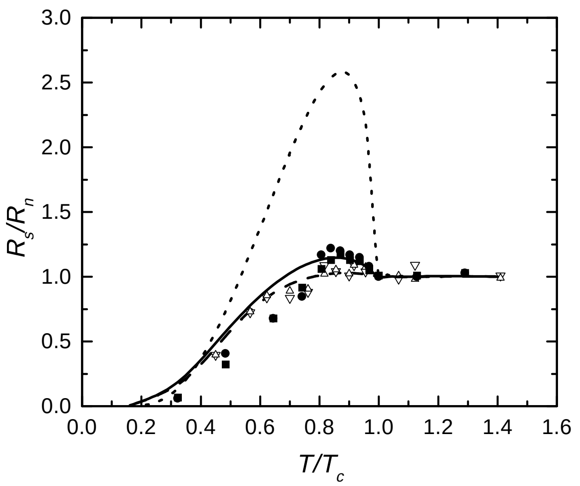

FIG. 2. The normalized relaxation rate, R_s/R_n, as a function of the reduced temperature T/T_c. The solid curve is for $UN_F = 0.31$ and $\lambda_s = 0.71$ corresponding to Fig. 1(a), the dotted curve is for the noninteracting case of $UN_F = 0$ and $\lambda_s = 0.35$ corresponding to Fig. 1(b), and the dashed curve is for $UN_F = 0.4$ and $\lambda_s = 0.8$. The substantial dampings in the superconducting state for the interacting cases suppress the NMR coherence peak as can clearly be seen by comparing the above curves. The filled circles and squares are the data from Stenger *et al.*, and the open up and down triangles are those from Sasaki *et al.* See the text for more detailed discussions.

against the theoretical curves in Fig. 2. The filled circles and squares represent, respectively, the data by Stenger *et al.* from ^{13}C NMR of Rb_2CsC_{60} at $B = 1.5$ T and $B = 3$ T, and the open up and down triangles represent the data by Sasaki *et al.* from ^{13}C NMR of K_3C_{60} at $B = 2.93$ T. It seems that the experimental data can be well described by $UN_F \approx 0.3-0.4$ and $\lambda_s \approx 0.7-0.8$. We note that Sasaki's data show a somewhat more suppressed NMR coherence peak compared with Stenger's data, while Stenger's data show a bit narrower width compared with the theoretical calculations.

We point out that Mazin *et al.* have analyzed the NMR data using the Eliashberg theory with two peaks in $\alpha^2F(\Omega)$, one high frequency peak at around 1000 cm^{-1} of corresponding $\lambda = 0.5$ and another very low frequency peak around 40 cm^{-1} of $\lambda = 2.7$ [19]. Such a low frequency mode with the strong coupling gives $\omega_{ln} = 66\ \text{cm}^{-1}$ in sharp contrast with the commonly held view of $\omega_{ln} \sim 1000\ \text{cm}^{-1}$. The present Letter explains the NMR coherence peak suppression without such a low frequency mode, and confirms that the BCS-Eliashberg framework of $\omega_{\text{ph}} \sim 1000\ \text{cm}^{-1}$ and $\lambda \sim 0.5-1$ can provide a consistent description of the wide range of experimental data including T_1, provided that the frequency dependent Coulomb interaction and the vertex correction are properly incorporated.

In summary, we have extended the standard Eliashberg theory for narrow bandwidth superconductors by including the frequency dependent screened Coulomb interaction together with the electron-phonon interaction, and by including the vertex correction via Nambu's method. We then solved the Eliashberg-Nambu equation self-consistently at all temperatures to obtain the gap and renormalization functions, $\Delta(\omega)$ and $Z(\omega)$, respectively, which are used to calculate the nuclear spin-lattice relaxation rate, T_1^{-1}. The frequency dependent Coulomb interactions between conduction electrons induce the substantial dampings in the superconducting state and, consequently, suppress the NMR coherence peak in the fullerene superconductors. The present Letter, therefore, has shown that the T_1 experiments can be understood with the view of $\omega_{\text{ph}} \sim 1000\ \text{cm}^{-1}$ and $\lambda \sim 0.5-1$. It remains to be seen if other experimental data can also be understood with the view.

The author thanks Dr. Charles Pennington and Dr. Susumu Sasaki for useful discussions, and acknowledges the support by Korea Science and Engineering Foundation (KOSEF) through Grant No. 96-0702-02-01-3, through the Center for Theoretical Physics, Seoul National University, and by the Ministry of Education through Grant No. BSRI-97-2428.

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