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LETTER TO THE EDITOR

Reduction of the recoilless fraction of the Mössbauer spectra in superconductors

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Abstract. We find that the recoilless fraction of the Mössbauer spectrum is reduced in the superconducting state because of the reduction in the single-phonon frequency due to the electron pairing. The reduction in the recoilless fraction depends on the ratio of the square of the gap energy to the attractive energy. The predicted reduction is in accord with the measurement of the Mössbauer spectra of ^{57}Fe in the $Bi_4Sr_3Ca_3Cu_{3.92}Fe_{0.08}O_{16.36}$ superconductor.

The intensity of the Mössbauer line is determined by the recoilless fraction

$$f = \exp\left(-\tfrac{3}{4}(\hbar^2 k_\gamma^2 / M k_B \theta_D)\right) \tag{1}$$

where the exponent contains the ratio of the recoil energy to the Debye energy $k_B\theta_D$. Here M is the mass per atom, k_γ is the wavevector of the γ-ray and θ_D is the Debye cutoff temperature. It may be written as

$$f = \exp\left(-\tfrac{1}{3}k_\gamma^2\langle u^2\rangle\right) \tag{2}$$

where $\langle u^2\rangle$ is the mean-square distance between atoms [1].

In this letter, we wish to calculate the effect of optical phonons on the recoilless fraction of the Mössbauer spectrum in a superconductor. We use a BCS-type theory to find the change in the single-phonon energy which we use to determine the number density of the phonons in the paired state. Since the single-particle energy decreases, the number density increases so that the mean square displacement of an atom, $\langle u^2\rangle$, increases, leading to a decrease in the f-factor of the γ-ray in going from the normal to the paired state.

We consider the interaction of conduction electrons with lattice phonons so that the Hamiltonian becomes

$$H = \sum_{k,\sigma} E_{k,\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_q \hbar\omega_q a_q^\dagger a_q + \sum_{k,q,\sigma} D_q c_{k+q,\sigma}^\dagger c_{k,\sigma} a_q + \sum_{k,q,\sigma} D_{-q} c_{k-q,\sigma}^\dagger c_{k,\sigma} a_q^\dagger + \text{h.c.} \tag{3}$$

where D_q is the matrix element of the electron–phonon interaction and h.c. stands for the hermitean conjugate of the previous terms. The operators $c_{k\sigma}^\dagger(c_{k,\sigma})$ create (destroy) electrons in the conduction band and $a_q^\dagger(a_q)$ create (destroy) phonons. $E_{k\sigma}$ is the single-electron energy and $\hbar\omega_q$ is the single-particle phonon energy. Using the anticommutators for electrons and commutators for phonons, the electron–phonon interaction transformed up to second order may be written as

$$H' = \tfrac{1}{2}\sum_{k,k',\sigma,\sigma'} V_{k,k'} c_{k',\sigma'}^\dagger c_{-k',\sigma}^\dagger c_{-k,\sigma} c_{k,\sigma'} \tag{4}$$

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where

$$V_{k,k'} = 2|D_{k-k'}|^2\hbar\omega_{k-k'}[(E_k - E_{k'})^2 - (\hbar\omega_{k-k'})^2]^{-1}. \tag{5}$$

We introduce the gap as in the BCS theory [2], as

$$\Delta_k = \sum_{k'} V_{k,k'} c_{k'}^{\dagger} c_{-k'}^{\dagger} \tag{6}$$

where k is associated with spin up while $-k$ is with spin down so that the gap corresponds to spin singlet state. Substituting (6) into (4) we find

$$H' = \tfrac{1}{2}\sum_{k,\sigma,\sigma'} \Delta_{k'} c_{-k,\sigma} c_{k,\sigma'} + \tfrac{1}{2}\sum_{k',\sigma,\sigma'} \Delta_{k'} c_{k',\sigma'}^{\dagger} c_{-k',\sigma}^{\dagger} + \frac{1}{2V_{k,k'}}\Delta_k \Delta_{k'}. \tag{7}$$

The potential (5) is attractive for $E_k - E_{k'} \ll \hbar\omega_{k-k'}$, otherwise it is repulsive. The attractive potential is

$$v_{k-k'} = -2|D_{k-k'}|^2/\hbar\omega_{k-k'}. \tag{8}$$

The average value of the interaction (7) is thus found to be

$$\langle H' \rangle = -2\Delta^2/v_{k-k'} \tag{9}$$

where $\Delta^2 = \Delta_k \Delta_{-k}$, which means that the square of the gap energy is used. Alternatively, we calculate the second-order energy of the system from the Hamiltonian (3) from which we evaluate the derivative with respect to the phonon number density to determine the self energy of the phonon which upon the use of pairing operator gives the modified single particle energy of the phonon as

$$\hbar\omega' = \hbar\omega_q - 2\Delta^2/v_q \tag{10}$$

which shows that the frequency of the phonons is reduced upon pairing by the amount $-2\Delta^2/v_q = -(\Delta^2/D_q^2)\hbar\omega$. The phonon number density then becomes

$$n_q = (\exp((\hbar\omega_q - 2\Delta^2/v_q)/k_B T) - 1)^{-1} \tag{11}$$

so that the average value of the square of the atomic displacement becomes

$$\langle u^2 \rangle = \frac{3(2\pi)^{1-d}\hbar}{\rho v^d}\left(\frac{k_B T}{\hbar}\right)^{d-1} \int (2n+1)x^{d-2}\, \mathrm{d}x \tag{12}$$

where ρ is the mass density of the solid, v the sound velocity and d the dimensionality [3]. For $d = 3$, it has two terms, a zero-point contribution, $u^2(0)$ and a temperature-dependent term, $\langle u^2(T) \rangle$, as

$$\langle u^2 \rangle = \langle u^2(0) \rangle + \langle u^2(T) \rangle \tag{13a}$$

with

$$\langle u^2(0) \rangle = \frac{3\hbar}{2(2\pi)^2\rho v^3}\left(\frac{k_B \theta_D}{\hbar}\right)^2 \tag{13b}$$

$$\langle u^2(T) \rangle = \frac{3}{2\pi^2\rho v^3}\int [\exp\{(\hbar\omega_q - 2\Delta^2/v_q)/k_B T\} - 1]^{-1}(\hbar\omega_q - 2\Delta^2/v_q)\, \mathrm{d}\omega_q. \tag{13c}$$

In order to understand the integral further, we need the wavevector dependence of v_q. For optical phonons, we assume

$$D_q = D_0(\hbar/2M\omega_q)^{1/2} \tag{14}$$

where $D_0 = (\partial V/\partial r)_0$ is the distance derivative of the crystal potential evaluated at the equilibrium. We define a parameter

$$s_0 = \Delta^2/(D_0^2\hbar^2/2M) \tag{15}$$

so that using (8) and (14), the temperature dependent part of the amplitude of the oscillation becomes

$$\langle u^2(T)\rangle = \frac{3}{2\pi^2\rho v^3}\int (\exp((\hbar\omega_q - s_0\hbar^2\omega_q^2)/k_BT) - 1)^{-1}(\hbar\omega_q - 2\Delta^2/v_q)\,d\omega_q \tag{16}$$

Substituting $\hbar\omega/k_BT = x$, we find that

$$\langle u^2(T)\rangle = \frac{3\hbar}{2\pi^2\rho v^3}\left(\frac{k_BT}{\hbar}\right)^2\int (\exp(x - s_0x^2k_BT) - 1)^{-1}x\,dx$$
$$-\frac{3s_0\hbar^2}{2\pi^2\rho v^3}\left(\frac{k_BT}{\hbar}\right)^3\int (\exp(x - s_0x^2k_BT) - 1)^{-1}x^2\,dx \tag{17}$$

which describes the reduction of the logarithm of the f factor upon pairing. The larger the gap energy, the larger is the reduction. Upon cooling, further reduction is predicted.

In the case of ^{119}Sn in $EuBa_2Cu_{2.98}Sn_{0.02}O_{7-\delta}$, the value of f at $T = 0$ extrapolated from the normal state is about 0.868 whereas the actually measured value [3] in the superconducting state is 0.800. This reduction of 0.068 amounts to about 8.5% in going from the normal to the superconducting state. Our calculation shows that the recoil-free fraction is indeed reduced in going from the normal to the superconducting state by about this amount. There is a large reduction [4] in the recoilless fraction of ^{57}Fe in $YBa_2(Cu_{0.98}Fe_{0.02})_4O_8$ superconductor but the data are masked by a structural distortion. However, we have found [5] that the isomer shift in this system is correlated with the diamagnetic susceptibility.

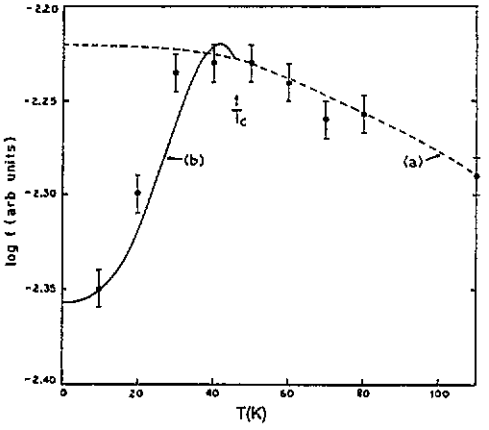

Figure 1. The recoilless fraction of ^{57}Fe in $Bi_4Sr_3Ca_3Cu_{3.92}Fe_{0.08}O_{16.36}$ (a) in the normal state and (b) in the superconducting state. The curves are calculated as given in the text and the dots are the values of the logarithm of the absorption area experimentally measured by Lin and Lin [6].

The integrals in (17) at temperatures lower than the Debye cutoff temperature are independent of temperature. Therefore, the logarithm of the recoilless fraction varies as

$$-\tfrac{1}{3}k_\gamma^2\langle u^2\rangle = -a_1T^2 + b_1T^3(1 - (T/T_c)) + c \tag{18}$$

where

$$a_1 = \frac{k_B^2(k_\gamma^2/3)}{4\rho v^3\hbar} \tag{19a}$$

$$b_0 = \frac{6Mk_B^3\zeta(3)(k_\gamma^2/3)}{\pi^2\rho v^3\hbar^3D_0^2} \qquad b_1 = b_0\Delta_0^2. \tag{19b}$$

The quantity c is caused by the quantum mechanical corrections such as those arising from the first term of (13a). Here, $\zeta(3)$ is the Riemann zeta function and we have used the temperature-dependent gap

$$\Delta^2 = \Delta_0^2(1 - T/T_c) \tag{20}$$

for the BCS singlet state. This equation (18) describes a peaked function with the value at T_c deeper than at $T = 0$. Such a dependence is not comparable with the experimental data [6] of the recoilless fraction of ^{57}Fe in $Bi_4Sr_3Ca_3Cu_{3.92}Fe_{0.08}O_{16.36}$. For this purpose we use a complex or a triplet type gap

$$\Delta^2 = \Delta_1^2 + \Delta_0^2(1 - T/T_c) \tag{21}$$

where Δ_1 may correspond to a gap energy corresponding to structural distortion whereas Δ_0 corresponds to the superconducting gap. The structural distortions have been detected by Boolchand *et al* [3]. The result (18) with structural distortions may thus be written in the form

$$-\tfrac{1}{3}k_\gamma^2\langle u^2\rangle = -a_1T^2 + b_1T^3(1 - T/T_c) + b_2T^3 + c \tag{22}$$

where $b_2 = b_0\Delta_1^2$. In figure 1, we show the predicted value of the logarithm of the recoilless fraction of ^{57}Fe in $Bi_4Sr_3Ca_3Cu_{3.92}Fe_{0.08}O_{16.36}$ in arbitrary units, for $a_1 = 5 \times 10^{-6}$ K^{-2}, $b_1 = 6.4 \times 10^{-6}$ K^{-3}, $b_2 = 1.4134 \times 10^{-6}$ K^{-3}, $c = -2.357$ and $T_c = 46$ K. The normal state value is also shown. We find that the predicted temperature dependence of the recoilless fraction in the superconducting state is in reasonable agreement with the experimental data only with the complex gap as in (21). The resistivity of the sample drops to 50% at 53 K and becomes zero at $T_c' = 33$ K. Thus our value of $T_c \simeq 46$ K is in accord with the value at which the resistivity is about one half of the normal state value. From the measured value of a_1 and the expression (19*a*) we measure the velocity of sound. Using the known values of the mass density, $\rho \simeq 6$ g cm^{-3} and the γ-ray wavevector $k_\gamma = 7.29 \times 10^8$ cm^{-1} corresponding to $E_\gamma \simeq 14.4$ keV, we calculate $v = 2.98 \times 10^5$ cm s^{-1} for the superconductor. This value of the velocity of sound in a pellet is quite reasonable. Thus the recoilless fraction is used as a method of measuring the sound velocity. The parameters b_1 and b_2 are dependent on a_1 and on the ratio of the gap energy to the matrix element of the electron–phonon interaction. To demonstrate this aspect of the theory we write

$$b_1 = (12\zeta(3)a_1\delta_c^2)/(\pi^2T_c) \tag{23}$$

where

$$\delta_c = \Delta_0/(\hbar D_0/(2Mk_BT_c)^{1/2}) \tag{24}$$

For the measured values of a_1, b_1, T_c and $\zeta(3) = 1.2$, we calculate $\delta_c = 6.35$ which shows that the strength of the electron–phonon interaction is about one sixth of the gap energy which is quite reasonable for the BCS theory. Since the structural-distortion temperature is very near the transition temperature, we expect b_1 and b_2 to be of the same order of magnitude as they are. The value of Δ_0 derived from the measured value of b_1 is in accord with the BCS relation $2\Delta_0 = 3.5k_BT_c$ for the given value of δ_c.

In conclusion, we find that the recoilless fraction of a Mössbauer line is reduced in going from the normal to the superconducting state. The reduction is related to the gap energy of the electron dispersion relation. The recoilless fraction in a superconducting lattice is thus obtained here from the BCS theory for the first time.

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