Isotope shift of the ferromagnetic transition temperature in itinerant ferromagnets

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ABSTRACT

We present a theory of the isotope effect of the Curie temperature T_c in itinerant ferromagnets. The isotope effect in ferromagnets occurs via the electron–phonon vertex correction and the effective attractive interaction mediated by the electron–phonon interaction. The decrease of the Debye frequency increases the relative strength of the Coulomb interaction, which results in a positive isotope shift of T_c when the mass M of an atom increases. Following this picture, we evaluate the isotope effect of T_c by using the Stoner theory and a spin-fluctuation theory. When T_c is large enough as large as or more than 100 K, the isotope effect on T_c can be measurable. Recently, precise measurements on the oxygen isotope effect on T_c have been performed for itinerant ferromagnet $SrRuO_3$ with $T_c \sim 160$ K. A clear isotope effect has been observed with the positive shift of $T_c \sim 1$ K by isotope substitution ($^{16}O \rightarrow {}^{18}O$). This experimental result is consistent with our theory.

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1. Introduction

Strongly correlated electron systems (SCES) have been investigated intensively, because SCES exhibit many interesting quantum phenomena. SCES include, for example, cuprate high-temperature superconductors [1–4], heavy fermions [5–8], and organic conductors [9]. In the study of magnetism, the Hubbard model is regarded as one of the most fundamental models [10–17]. The electron–phonon interaction is also important in metals and even in correlated electron systems. The electron–phonon interaction has a ubiquitous presence in materials.

The isotope effect of the ferromagnetic transition has been investigated for several materials. They are $La_{1-x}Ca_xMnO_3$ [18,19], $Pr_{1-x}Ca_xMnO_3$ [20], $RuSr_2GdCu_2$ [21], $ZrZn_2$ [22] and $SrRuO_3$ [23]. First three compounds $La_{1-x}Ca_xMnO_3$, $Pr_{1-x}Ca_xMnO_3$ and $RuSr_2GdCu_2$ show that T_c decreases upon the isotope substitution $^{16}O \rightarrow {}^{18}O$. The isotope shift of T_c for $ZrZn_2$ was not determined because the shift of T_c is very small and there was uncertainty arising from different impurity levels. The compound $SrRuO_3$ exhibits a positive isotope shift, that is, T_c increases upon ^{18}O isotope substitution. We think that mechanisms of the isotope effect for the first three materials and the last one $SrRuO_3$ are different.

The large Curie temperature shift $T_c(^{16}O) = 222.7$ K to $T_c(^{18}O) = 202.0$ K was reported when $x = 0.20$ for $La_{1-x}Ca_xMnO_3$ [18,19]. We consider that this shift is caused by strong electron-lattice coupling with some relation to large magnetoresistance [24,25]. There is a suggestion that the ferromagnetic transition is caused by the double-exchange interaction [26–28] and a strong electron-lattice interaction originating from the Jahn–Teller effect [29]. $Pr_{1-x}Ca_xMnO_3$ is also a member of materials that exhibit the colossal magnetoresistance phenomenon [20]. The Curie temperature was lowered due to the isotope substitution $^{16}O \rightarrow {}^{18}O$; $T_c(^{16}O) = 112$ K is shifted to $T_c(^{18}O) = 106$ K when $x = 0.2$. It is expected that the isotope effect arises from the same mechanism as for $La_{1-x}Ca_xMnO_3$ [30,31].

As for strontium ruthenates, Raman spectra of $SrRuO_3$ films showed anomalous temperature dependence near the ferromagnetic transition temperature [32]. This indicates that the electron–phonon interaction plays a role in $SrRuO_3$. Recently, the isotope effect of the Curie temperature T_c has been reported in $SrRuO_3$ [23]. This material is an itinerant ferromagnet with $T_c \simeq 160$ K. The ferromagnetic transition temperature was increased about 1 K upon ^{18}O isotope substitution. A softening of the oxygen vibration modes is induced by the isotope substitution ($^{16}O \rightarrow {}^{18}O$). This was clearly indicated by Raman spectroscopy. The Raman spectroscopy also confirmed that almost all the oxygen atoms (more than 80 percent) were substituted successfully. The increase of the atomic mass leads to a decrease of the Debye frequency ω_D. In

fact, the Raman spectra clearly indicate that the main vibration frequency of ^{16}O at 372 cm^{-1} is lowered to 351 cm^{-1} for ^{18}O by oxygen isotope substitution in SrRuO$_3$. This shift is consistent with the formula $\omega_D \propto 1/\sqrt{M}$ where M is the mass of an oxygen atom. Thus, experiments confirmed that the isotope shift of T_c is induced by the decrease of the frequency of the oxygen vibration mode.

In this paper we investigate the isotope shift of the Curie temperature theoretically. The paper is organized as follows. In the next Section, we outline the theory of isotope effect in a ferromagnet. In the Section 3 we show the Hamiltonian. In the Section 4, we examine the corrections to the ferromagnetic state due to the electron–phonon interaction, by examining the ladder, self-energy and vertex corrections. In the Section 5, we calculate the oxygen-isotope shift of T_c on the basis of the spin-fluctuation theory. We show that the both theories give consistent results on the isotope effect.

2. Isotope effect in a ferromagnet

The reduction of the Debye frequency results in the increase of relative strength of the Coulomb interaction U. This results in a positive isotope shift of T_c. This is a picture that indicates the positive isotope shift of T_c; $\partial T_c / \partial M > 0$.

We start from the Hubbard model with the on-site Coulomb repulsion U to describe a ferromagnetic state. The Curie temperature T_c is determined by the gap equation. The effective attractive interaction due to the phonon exchange reduces U to $U + g$ ($g < 0$) in the neighborhood of the Fermi surface. The effective attraction, however, shows no isotope shift in the Stoner theory because the Curie temperature is determined by the interaction at the Fermi surface and then the variation of ω_D has no effect on T_c. The electron–phonon vertex correction reduces the magnetization and this leads to the isotope effect. Although the vertex correction is on order of ω_D / ϵ_F, for the Debye frequency ω_D and the Fermi energy ϵ_F, the isotope effect can be observed by precise measurements when the Curie temperature is as large as 100 K or more than that.

The isotope effect in itinerant ferromagnets was first investigated on the basis of the Stoner theory in Ref. [33], and the formula for isotope coefficient α was given. A fluctuation effect, however, is not included in the Stoner theory. Because the spin-fluctuation theory has been successful in understanding physical properties in itinerant ferromagnets [10], a formula based on the spin-fluctuation theory is necessary. We present the formula of the isotope coefficient on the basis of the spin-fluctuation theory, and show that the isotope effect observed by experiments is consistent with this formula.

3. Hamiltonian

The total Hamiltonian is the sum of the electronic part, the phonon part and the electron–phonon interaction part:

$$H = H_{el} + H_{ph} + H_{el-ph}. \tag{1}$$

Each term in the Hamiltonian is given as follows.

We adopt that the ferromagnetism arises from the on-site Coulomb interaction and use the Hubbard model given as

$$H_{el} = \sum_{\mathbf{k}\sigma} \xi_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \tag{2}$$

where $c_{\mathbf{k}\sigma}$ and $c_{\mathbf{k}\sigma}^\dagger$ are Fourier transforms of the annihilation and creation operators $c_{i\sigma}$ and $c_{i\sigma}^\dagger$ at the site i, respectively. $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is the number operator, and U is the strength of the on-site Coulomb interaction. $\xi_{\mathbf{k}} = \epsilon_{\mathbf{k}} - \mu$ is the dispersion relation
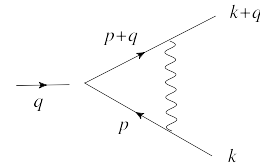
Fig. 1. Electron–phonon vertex function. The wavy line indicates the phonon propagator. The momenta k, p and q represent 4-momenta such as $k = (i\epsilon_m, \mathbf{q})$, $p = (i\omega_n, \mathbf{p})$ and $q = (i\nu_\ell, \mathbf{q})$.

measured from the chemical potential μ. The phonon part of the Hamiltonian is given by

$$H_{ph} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \frac{1}{2} \right), \tag{3}$$

where $b_{\mathbf{k}}$ and $b_{\mathbf{k}}^\dagger$ are operators for the phonon and $\omega_{\mathbf{k}}$ is the phonon dispersion. The electron–phonon interaction is [34]

$$H_{el-ph} = \gamma \int d^3 x \sum_\sigma \psi_\sigma^\dagger(\mathbf{x}) \psi_\sigma(\mathbf{x}) \varphi(\mathbf{x}), \tag{4}$$

where the electron field ψ_σ and the phonon field φ are defined, respectively, as follows:

$$\psi_\sigma(\mathbf{x}) = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}} c_{\mathbf{k}\sigma}, \tag{5}$$

$$\varphi(\mathbf{x}) = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \left(\frac{\hbar\omega_k}{2} \right)^{1/2} \left(b_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}} + b_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{x}} \right), \tag{6}$$

where V is the volume of the system.

4. Electron–phonon vertex correction

4.1. Electron–phonon vertex function

The electron–phonon vertex correction plays an important role in the isotope effect in itinerant ferromagnets. The vertex function $\Gamma(k, q + k; q)$, shown in Fig. 1, is written as

$$\Gamma(k, k+q; q) = -\gamma^2 \frac{1}{\beta} \sum_n \int \frac{d^3 p}{(2\pi)^3} G_0(i\omega_n, \mathbf{p})$$
$$\times G_0(i\omega_n + i\nu_\ell, \mathbf{p} + \mathbf{q}) D_0(i\omega_n - i\epsilon_m, \mathbf{p} - \mathbf{k}), \tag{7}$$

where G_0 is the electron Green function and D_0 is the phonon Green function [34]:

$$G_0(i\omega_n, \mathbf{p}) = \frac{1}{i\omega_n - \xi_{\mathbf{p}}}, \tag{8}$$

$$D_0(i\nu_\ell, \mathbf{k}) = \frac{\omega_{\mathbf{k}}^2}{(i\nu_\ell)^2 - \omega_{\mathbf{k}}^2}, \tag{9}$$

where $\omega_{\mathbf{k}}$ is the phonon dispersion relation. It is known as the Migdal theorem that the vertex correction is of order of ω_D / ϵ_F [34–38]. The vertex function is evaluated by using the method of Green function theory [34,38,39]. In the limit $(i\nu, \mathbf{q}) \to 0$, we obtain [38]

$$\Gamma(k, k+q; q) \simeq -\gamma^2 \rho(0) \frac{1}{2} \frac{\omega_D}{\epsilon_F} \ln\left(\frac{\epsilon_F}{\omega_D} \right). \tag{10}$$

We consider the self-energy corrections shown in Fig. 2. The first term is the Hartree term that stems from the on-site Coulomb interaction and the second one includes the vertex correction. When we use the approximation in eq. (10), the self-energy is