Phonon echoes in Si:P at very low temperature

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Abstract. We observed phonon echoes in P-doped Si (Si:P) at very low temperatures. We applied two radio-frequency pulses separated by a time delay of \(\tau\) on Si:P and observed echo signal at \(t = 2\tau\) in both insulating and metallic samples with varying dopant concentrations and of different sample forms of powders and bulk plates at temperature between 45 mK and 4 K. The echoes were much more pronounced in insulating powder samples than in metallic ones and in bulk ones. The echo intensity for a fixed \(\tau\) increased very strongly as temperature was lowered but the echoes disappeared toward the superfluid-to-normal transition temperature of helium mixture in which the samples were immersed. We observed no appreciable change in the echo intensities as external magnetic field was varied up to 8 T. The echoes are interpreted to be dynamical polarization phonon echoes in piezoelectric powders of insulating Si:P with a dopant concentration \(n = 6 \times 10^{17} \text{ cm}^{-3}\).

1. Introduction

Echo phenomena are found in various physical systems. Most well-known ones are spin echoes observed when the radio-frequency or microwave pulses are applied on spin systems [1]. There are photon echoes from optical transition in materials [2, 3], cyclotron echoes in plasma [4] and so on. One interesting class of echo phenomena, broadly termed as phonon echoes or polarization echoes, include those which are mostly observed in powders of piezoelectric materials [5, 6], metals [7, 8], superconductors [7, 9], glasses [10], etc.

We report the unusual features of phonon echoes found in insulating powder sample of phosphorus-doped silicon (Si:P). The echoes were found at very low temperatures below 2 K in the course of \(^{31}\)P-nuclear magnetic resonance (NMR) experiment. The echoes almost disappeared as the temperature approaches the normal-to-superfluid transition temperature of the helium mixture (5% \(^{3}\)He concentration of the mixture) where the sample was immersed. The echoes did not depend on applied magnetic field up to 8 T.

2. Experiments and Results

We prepared Si:P samples with various dopant concentrations. We investigated three samples, insulating powder and 0.2 mm-thick plates with \(n = 6 \times 10^{17} \text{ cm}^{-3}\) and metallic powder with \(n = 6 \times 10^{19} \text{ cm}^{-3}\) where the critical P-dopant concentration for the metal-insulator transition
\( n_c = 3.7 \times 10^{18} \text{ cm}^{-3} \). The samples were immersed in the \(^3\text{He} \) and \(^4\text{He} \) mixture to cool down to about 40 mK by a dilution refrigerator. We applied two rf pulses (100 ∼ 120 MHz) with time delay \( \tau \) separating the first and second pulses of the sequence of \( \pi/2 - \tau - \pi \), where \( \pi/2 \) and \( \pi \) do not represent the rotation angles of magnetization in the Bloch sphere but just the relative pulse lengths (\( \pi/2 : 2\mu\text{s} \) and \( \pi : 4\mu\text{s} \)). The echo signal observed in the insulating powder sample was bigger by one or two order of magnitude than in metallic sample and in bulk plates. The large echo observed in insulating sample compared to the tiny signal in metallic one suggests that these echoes are of different origin from those found in metallic powders \([7, 8]\) and the surface effect may not be the cause for echoes. Sound may be resonated with the Si samples with 10 \( \mu \text{m} \) in size in 100 MHz frequency range and thus the powder sample is better coupled to rf-field pulse than the plates. We focus our discussion on data of the insulating powder sample with the dopant concentration \( n_c = 6 \times 10^{17} \text{ cm}^{-3} \).

First, we investigated the magnetic field dependence of the echo at a fixed \( 2\tau = 150\mu\text{s} \). We varied the external field from 8 T down to no-field and observed no field dependence as shown in Fig. 1. This observation allows us to exclude any mechanisms of magnetic origins such as spins or magneto-acoustic modes \([7]\). The echo intensity strongly depends on temperatures.

We obtained the characteristic time constants \( T_2 \) for the echoes. Figure 2 shows the echo amplitudes as \( 2\tau \) varies. The time constant obtained from the slope of the echo envelope is called \( T_2 \) in general and corresponds to the time scale of the destructive interference of the phases of precessing spins in magnetic resonance. In polarization phonon echo experiment, however, \( T_2 \) corresponds to the energy dissipation and the meaning of which might be closer to \( T_1 \) in a conventional term. The initial buildup and the following decay of the echo amplitude as a function of \( 2\tau \) as shown in Fig. 2 is characteristic for the model of the anharmonic oscillators which can be described by the following equation,

\[
\ddot{S} - 2\Gamma \dot{S} + \omega_0^2 \left(1 - \gamma S^2\right) S = f(t),
\]

where \( S \) is the strain, \( \Gamma = 1/T_2 \) the frictional loss, \( \gamma \) corresponds to the nonlinearity of elastic stiffness, and \( f(t) \) the perturbation such as rf pulses which couples to the acoustic oscillation. The echo amplitude as a function of \( 2\tau \), \( E(2\tau) \), obtained from the solution to the above equation is given as,
Figure 2. The echo intensities with varying the time delay \( \tau \) between the two pulses.

\[
E(2\tau) \sim \frac{T_2}{2} \gamma E_1 E_2^2 (1 - e^{-2\tau/T_2}) e^{-2\tau/T_2},
\]

where \( E_1 \) and \( E_2 \) are the sound impulses induced by the 1st and 2nd rf-pulses, respectively. The echo builds up in a time of \( T_2 \) and decays with a time constant of \( T_2 \), that is characteristic for the dynamic polarization echoes. In Fig. 2, we fit \( E(2\tau) \) at \( T = 150 \) mK to this solution with \( \gamma E_1 E_2^2 \) and \( T_2 \) as the fitting parameters. We assume that only \( T_2 \) in Eq.(2) depends on temperatures and the high temperature data at \( T = 500 \) mK were fitted with \( T_2 \) as the fitting parameter with the same value of \( \gamma E_1 E_2^2 \) obtained from data at \( T = 150 \) mK. The results are lines in the Fig. 2. We obtained \( T_2 \simeq 2 \) ms and 1.2 ms for 150 mK and 500 mK, each respectively.

3. Analysis of \( T_2 \) and Discussion

Figure 3(a) shows the echo intensities observed at fixed \( 2\tau = 150 \) \( \mu \)s as a function of temperatures. We note the temperature where the echoes disappear corresponds to the superfluid-to-normal transition temperature of the mixture where the sample was immersed. The superfluidity of the mixture of liquid helium is treated as two fluid model [11] and the dissipative term in Eq.(1) is attributed due to quasi particle collision of the normal fluid component of the mixture to the particle. Since the mean free path becomes longer than the viscous penetration depth, the collision of the quasi particle can be treated in the Knudsen regime (ballistic limit) at low temperatures and \( 1/T_2 \) is given by,

\[
\left( \frac{2}{T_2} \right) = \frac{A \rho_n v_n}{a \rho},
\]

where \( A \) is the geometrical factor of order 1 to be determined, \( a \) is the particle diameter, and \( \rho_n \) the normal fluid density for the mixture and \( \rho \) is the density of Si. At very low temperatures below 1 K, the normal fluid density in the mixture should be replaced by \( n_3 \times m_3^* \), where \( m_3^* \) is \(^3\text{He} \) effective mass in the mixture and \( v_n \) by \( v_F \) the Fermi velocity.

Figure 3(b) shows \( 1/T_2 \) and \( \rho_n \) vs. temperatures, plotted in log-scales. The only temperature dependent term in Eq.(3) is \( \rho_n \) and thus temperature dependence of \( 1/T_2 \) should be directly compared with \( \rho_n \). Unfortunately the measured values of \( \rho_n \) are limited at high temperatures [11]. Below 400 mK, \( 1/T_2 \) decreases. We speculate that the Fermi temperature of the mixture is 300 mK and \( \rho_n \) starts to be Fermi-degenerate and effective \(^3\text{He} \) density decreases linearly to temperature below the Fermi temperature.
The microscopic origin for the formation of dynamic polarization echoes comes from the nonlinear term of elastic stiffness. The coupling of the Si:P with rf-pulse comes from piezoelectricity of S:P sample. Pure silicon cannot be piezoelectric because the crystal structure has no inversion symmetry. We suspect phosphorus impurities play a role for it. In the metallic sample, electron wave function is delocalized and can not contribute to piezoelectricity. On the other hand, the electronic wave function of isolated donor has spherical symmetry and thus may not change the situation. When the dopant concentrations increases near $n_c$, we found large number of clusters of P-impurities in our ESR measurement in the insulating sample [12]. The clusters of P-dopant formed in the sample near $n_c$ may be the cause of piezoelectricity of Si:P.

4. Conclusion

We found phonon echoes in insulating Si:P powder at low temperature. The echoes are interpreted as the dynamic polarization phonon echoes. The echo intensity decreased as temperature increased and eventually disappeared near the superfluid-to-normal transition temperature. We obtained the temperature dependence of $T_2$. The dissipation mechanism of the phonon echoes are attributed due to collision of the normal fluid component to powder of the sample.

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